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CONDITIONING AND DETAILED ANALYSIS OF BIOMASS DERIVED FUEL GAS

ONGOING AND PLANNED WORK BY BATTELLE

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ABSTRACT

Gas turbine based power and cogeneration schemes are likely to become more favored as turbine efficiencies improve, but the economics of local power generation may depend on the use of low cost fuels other than natural gas. Opportunities may arise in the application of gas turbines in the pulp and paper industry and the wider use of biomass derived fuels in general.

These fuels, as produced, typically contain inorganic impurities originating from ash forming substances and other minor constituents of the feedstock. Also, depending on the biomass treatment process, they contain varying amounts of complex organic derivatives, commonly referred to as tars, and some simpler condensable vapors.

The Department of Energy is sponsoring work aimed at providing realistic data on low level constituents and impurities in gas derived by

indirect gasification of wood, some of which may have disproportionately severe effects on turbine operation, durability, and emissions performance. It is planned to sample gas from both laboratory scale (up to 20 tons/day) and pilot scale (200 tons/day) installations and to assess the effectiveness of wet scrubbing procedures and catalytic reforming of condensables in cleaning up the gases. This paper discusses the rationale for this work, experimental approach, and analytic procedures that will be used. The work will include the operation of a small (220-kWe) gas turbine to provide direct information on the impact of using the final biomass derived gas delivered by the system.

The laboratory scale work is currently under way, with a planned completion date in mid 2000. The second phase is dependent on arrangements for integration of the R&D effort with the operation of the pilot plant.

INTRODUCTION

The last two decades have seen a rapid increase in the use of land based gas turbines; to a large extent this has been a result of the availability of natural gas at low prices, which has in turn facilitated the development of high efficiency low cost turbines designed to capitalize on the high purity and convenience of this fuel. The U.S. Department of Energy (DOE) has encouraged these developments through its Advanced Turbine Systems (ATS) program; this program also recognizes the need to accommodate other fuels, including biomass, which has the special property of being renewable, with no net CO₂ production resulting from its use. However, biomass, in common with other solid fuels, generally must be converted to liquid or gaseous form before use in gas turbines and must be treated to remove impurities. In the case of gases derived from biomass, the impurities of special concern are metals (especially alkalis), certain gases (ammonia, hydrogen cyanide, hydrogen sulfide), and condensable vapors.

Condensables include water vapor and light hydrocarbons such as benzene and toluene as well as high molecular weight species, for example, naphthalene, anthracene, and pyrene, which commonly are referred to collectively as tars.

Under contract from DOE, Battelle has developed a novel indirectly fired biomass atmospheric pressure gasifier (Paisley and Overend 1995). This gasifier produces a gas of relatively high heating value (300-500 Btu/scf) that can be burned in gas turbines with only minor changes in fuel systems designed for natural gas. A typical gas composition is given in Table 1. Compared with gases produced by partial oxidation of biomass in air or oxygen, this gas has a higher hydrocarbon content; tar production is minimized by rapid heating and short gas residence time, (~ 1 second) in the gasifier but is still significant. Gas made in oxygen blown gasifiers is virtually free from hydrocarbons, but gases from air blown gasifiers operating at lower temperatures may contain some condensable hydrocarbons.

Trace metals inherent in biomass and in soil adhering to it contribute to ash carried with the raw product gas. Some sample analyses are shown in Table 2. Silicon is abundantly present in ash from monocot crops such as grasses, including corn and sugar (Nordin 1994) while alkali, particularly potassium, is found in varying amounts in all biomass ashes. Alkalis are a particular concern because of their propensity to form compounds of low melting point, which are potentially corrosive to metals and ceramics. Table 2 includes information on impurities found in biomass as harvested. The organic part of

biomass consists essentially of carbon, hydrogen and oxygen, but also contains minor amounts of nitrogen and sulfur. These may contribute to exhaust emissions, most importantly nitrogen oxides. Sulfur levels are usually too low to be of direct concern but may form corrosive salts with alkalis or other metals. It is important not to consider individual elements in isolation.

While the possible effects of solid and gaseous impurities on turbine operation are critical in technical terms, the economic and environmental acceptability of a total biomass energy concept may depend equally on side effects. In the case of the Battelle gasification system these include the effects of emissions from the gasifier combustor (which burns residual char) and effluents from the gas cleanup process. The raw products of relatively low temperature (below 700 C) pyrolysis of biomass contain notoriously complex organic substances, (see for example Pakdel, Couture, and Roy 1994). These

Table 1. Composition of Gas from Battelle Pilot Scale Gasifier – Typical Dry Analysis.

Constituent	Vol. Percent
H ₂	24.6
CO ₂	18.2
C ₂ H ₄	4.6
C ₂ H ₆	0.5
C ₂ H ₂	0.6
N ₂	8.4
CH ₄	11.5
CO	32.6

Source Paisley and Overend, 1995.

Table 2. Examples of Biomass Ash Compositions.

Specie	Ash, %	Ash Composition, % as harvested						
		Al	Ca	Fe	Mg	Na	K	Si
Bagasse	2-3	18	4.5	14.4	3.3	0.8	4.1	46.6
Corn Cob	2-3	-	0.8	0.1	8	0.6	40	50
Corn Stalk	5-10	6	14	1.4	18	20	0.1	40
Wheat Straw	10-15	1.7	10	1.9	2.5	1.2	25	46
Poplar	2-3	1	47	0.5	4.4	0.2	20	26
Pine	2-3	4.5	49	3.5	0.5	0.4	2.6	32.5
Maple	3-5	4	56	2	20	-	6	10

usually get only brief mention as "tars". In this paper, we define tars as hydrocarbon and oxygen containing hydrocarbons of molecular weight over 78. Between the gaseous products and tars there also are light condensables, principally benzene that may represent both a significant fuel value and a hazardous potential effluent. Overall, by control of heating rate, temperature, and reaction time, the Battelle process aims to recover the fuel values and eliminate effluents that might restrict its application.

The objective of the program described is to identify and quantify the impurities produced by the Battelle gasifier, and to evaluate the gas conditioning methods that are being applied. These include a catalytic treatment step in which tars are converted mainly to CO and H₂ prior to quenching in the scrubber. The evaluation process includes characterization of any waste streams that may be generated, and direct observation of fuel related performance changes in a Solar Spartan gas turbine operating on the product gas. Two phases are planned. The first one (which is already in progress) will be carried out using a 20 ton/day pilot scale gasifier at Battelle's Ohio site, the second (not currently funded) will use the same procedures at Burlington, Vermont, where a 200 ton/day demonstration gasifier has been installed and operated. Timing of the second phase is dependent on integration with the pilot plant program. The data generated will provide information for these sites as well as general implications on contaminants that may be experienced at any wood gasification plant, on clean up system performance, on sampling and analytic methods and on the effects on the turbine itself.

This paper is intended to provide information to encourage discussion on the adequacy of the measurement program and the requirements of gas turbines as they relate to biomass derived fuels.

PILOT PLANT

The essential features of the pilot plant are illustrated in Figure 1. There are four main subassemblies, namely:

- The feed system
- The gasifier, which includes both a gasifier shaft and a combustor vessel
- A gas conditioning and clean up section
- The gas compressor gas delivery system.

The gas turbine/generator is a separate self-contained package. The sampling points and gas turbine instrumentation are located to provide data on the performance of each of these subassemblies and to permit the preparation of energy and mass balances at appropriate interfaces.

Raw biomass is fed via a lock hopper system and a screw feeder into the base of the insulated stainless steel gasifier shaft, which is 0.25-m internal diameter and about 6-m high. In the gasifier, the feed is fluidized by steam, and meets a stream of hot inert solids from the combustor. Gas generated in the shaft accelerates the solids, which are carried into a cyclone separator and then to the combustor via a seal leg. In the combustor vessel, air is used to promote fluidization, to complete combustion of unreacted carbon, and to raise the temperature of the inert solids before they are returned to the gasifier. In the pilot plant, nitrogen is used to control the rate of this recycle flow via an L valve.

The measurement points in the feed system and gasifier subassembly are listed in Table 3, which refers to the locations indicated in Figure 1.

Gases leaving the combustor are exhausted to atmosphere after clean up and the separated solids may be returned to the combustor. Solids carried over with the product gas into the secondary cyclone are treated similarly.

Hot product gas may be conditioned in two ways. In the first, it passes directly through a conventional scrubber system, which incorporates a venturi scrubber section and a spray tower. In the second process, the hot product gas passes first through a fluidized hot catalyst bed in which the tars (which usually amount to about 1/2 percent molar of the total dry raw gas stream) are essentially eliminated by conversion to hydrogen, methane and carbon monoxide before passing to the scrubber. Preliminary results demonstrating this capability has been reported elsewhere (Paisley 1996).

The cooled gas leaving the scrubber is dewatered by pressure swing absorption and passes to the screw compressor. The gas delivery system incorporates a surge chamber to facilitate the changeover from natural gas to biogas after the gas turbine has been brought on line. The changeover is achieved on load and without any noticeable change in turbine operation.

The output of the laboratory gasifier is insufficient to support full load operation of the turbine. To permit gas turbine operation at full load and temperature, the biogas supply will be augmented by natural gas, which is premixed with the gasifier product before the compressor. At rated conditions, the feed gas consists of approximately 85 (volume) percent biogas and 15 percent natural gas. The main difference in composition is an increase in the methane content from 15 percent to 25 to 30 percent. The biogas supply rate is monitored prior to enrichment.

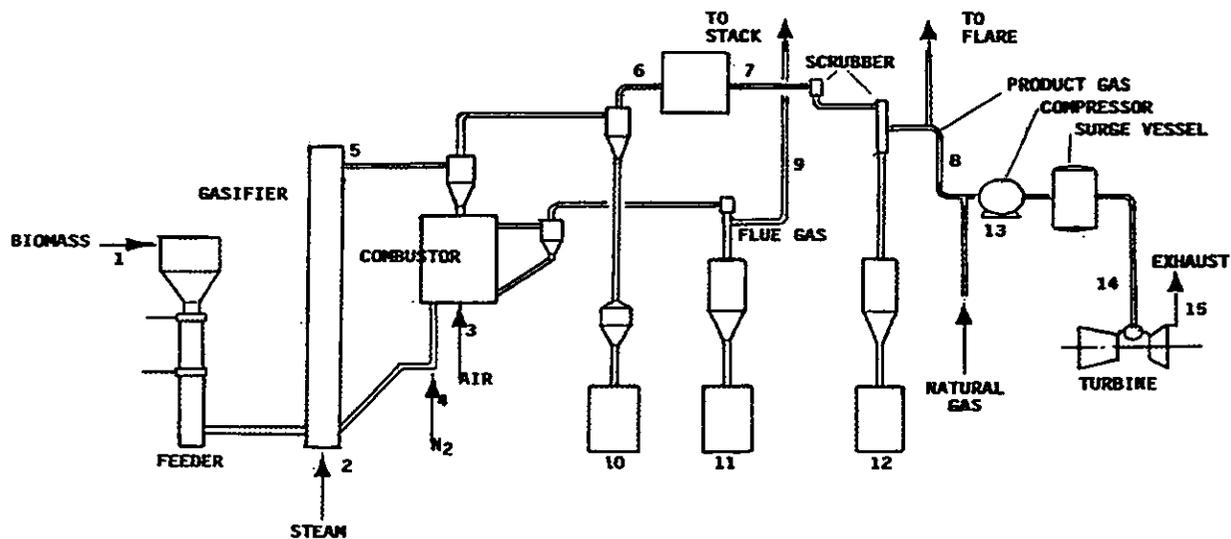


FIGURE 1. LABORATORY SCALE GASIFIER WITH SAMPLING LOCATIONS.

Table 3. Summary of Laboratory Unit Data Collection.

Sample Location	Data Collected	Frequency	
Feed System	1. Biomass input	Direct weighing and sample	Hour
	2. Fluidizing steam	Orifice flowmeter, T/C, P.	Cont.
	3. Combustion air	Orifice flowmeter, T/C, P.	Cont.
	4. N ₂ to L valve	Orifice flowmeter	Cont.
Outputs	5. Gasifier unit	Temp. T/C	Cont.
	6. Raw gas	CO, CO ₂ , H ₂ , CH ₄ , C ₂ , H ₂ O, Other organic (M.S.) Particulate, NH ₃	Cont. Cont. TBD
	7. Re-formed gas	CO, CO ₂ , H ₂ , CH ₄ , C ₂ , H ₂ O, Other organic (M.S.) Particulate NH ₃ , H ₂ S	Cont. Cont. TBD Cont.
	8. Product gas	CO, CO ₂ , H ₂ , CH ₄ , H ₂ O, Other organic (M.S.) Particulate NH ₃ , H ₂ S, condensables	Cont. Cont. TBD -
	9. Flue gas	CO, CO ₂ , O ₂ , SO _x , NO _x , UHC	Cont.
	10. Solids from raw gas	Direct weighing and sample	Hour
	11. Flue gas solids	Direct weighing and sample	Hour
	12. Scrubber effluent	Aqueous stream sample Organics sample Solids sample	Hour Hour Hour
	13. Compressor oil	Sample	8 Hours
	14. Gas to turbine	CO, CO ₂ , H ₂ , CH ₄ , C ₂	Cont.
	15. GT exhaust	O ₂ , NO _x , CO, CO ₂ , UHC, SO _x	Cont.

The objectives of the test phase described here require the collection of data on the gas composition and impurity levels leaving the gasifier, the gas conditioning system, the gas compressor, and the gas turbine. The compressor lubrication system also is monitored to quantify possible contamination from the gasifier product. Measurements made in the gasifier process section are important also in defining the origins of impurities and in establishing component mass balances.

Demonstration Plant

The flowsheet for the demonstration plant is very similar to that for the pilot plant, although the capacity is about ten times larger. The gasifier shaft in this case is a brick lined vessel 1.0-m in diameter and approximately 20.-m in height. The gasifier-fluidizing medium is steam. This plant does not incorporate a hot gas (catalyst) conditioning bed in the main process stream, but there will be one in the slip stream feeding the Solar Spartan turbine. The biogas supply will be sufficient without natural gas enrichment in this case. For the purposes of this program, the sampling and analysis methodology will concentrate on the gas entering and leaving the hot gas conditioning unit and entering the compressor, and on the turbine exhaust. The performance of the gasifier and the full stream gas cleanup system will be monitored as part of the gasifier demonstration program and will provide a basis to which the slip stream product can be compared. For further information on the demonstration unit see Paisley, et. al., 1997 (Reference 5).

Analytic Methods and Procedures

In general, as indicated in Table 3, the measurements follow accepted practice. CO and CO₂ Unburned Hydrocarbons (UHC) and SO₂ are measured by NDIR; NO_x by chemiluminescence; O₂ by a solid electrolyte cell; hydrocarbon gases by mass spectrometry (MS); tars and solid particulates are collected by a Modified EPA Method 5 for analysis by MS (for tars) or X-ray diffraction (XRD) and Inductively Coupled Plasma/MS (for solids).

Particulates in the gas stream are sampled isokinetically at the center of the flow stream and concentrations and size distributions measured by Andersen Impactor Analysis.

Solid particles collected from the gas stream will be analyzed by XRD to obtain information on both elements and associations or compounds that may be present. The collection of alkali compounds is difficult because some may be in vapor form at the higher temperatures and potentially can condense in the sampling train. Heated sampling lines will be used to minimize errors caused in this way.

Soluble salts and gases will be removed to varying degrees in the scrubber. Ideally, the aim is to account for all the solid forming inputs in the biomass supply by summation of the amounts found at the sampling locations. XRD will be augmented by absorption of the vaporized metals and fine particles in solutions that will be analyzed by Inductively Coupled Plasma/Mass Spectrometry. Past experience using these methods has shown that satisfactory mass balance closure can be obtained.

The most critical concerns involving solids are to define the efficiency of removal of solid contaminants in the scrubber system, the effects of the catalytic treatment bed, and the nature and concentrations of solids forming species entering the turbine. These data in turn will be compared with deposits found in the turbine after extended operation (~ 1000 hrs is planned) and in the turbine exhaust stream. In the event that significant deposits are found, samples will be taken from different temperature zones to gain a better understanding of deposition processes.

These standard test procedures will be augmented by performance testing of the gasifier and catalytic conditioning reactor using on-line, continuous, multi-species monitoring using NREL's Transportable Molecular Beam Mass Spectrometer (TMBMS) (Ratcliff, 1994). Previous monitoring experience with this instrument has shown that conventional continuous process measurements of carbon monoxide, carbon dioxide and methane concentrations give an incomplete picture of process chemistry and an unreliable indication of process stability (Ratcliff, et. Al., 1995). However, by the

addition of continuous TMBMS monitoring of syngas components including hydrogen, benzene, naphthalene, and phenol, much information about process chemistry and stability can be revealed. More than thirty species can be monitored simultaneously at rates as high as twice per second, although one per minute is probably adequate for this work. These species include benzene, which often is missed or inadequately represented in conventional analysis.

Process monitoring with the TMBMS provides time resolved, multidimensional, chemical data. These data may be used on-line to follow changes in the process, establish steady state operation and monitor tar decomposition efficiency. Thus, it can assist the plant operators in tuning the process for optimum performance and can probe the limits of throughput capacity and turndown ratio. Off-line, the data can be used to quantify many of the major gas and condensable species concentrations. The quantitative data will be useful in guiding the experimental program by providing chemical information from which decisions about operating condition changes, or catalyst/reactor modifications can be made.

In the laboratory installation at Battelle, the TMBMS together with conventional methods of analysis will monitor at the inlet and outlet of the hot gas-conditioning reactor. These samples will provide data to:

1. Determine raw product gas compositions as a function of time and process parameters including measurements of hydrogen, carbon dioxide and methane. Tar species including benzene, toluene, phenol, naphthalene, etc. will also be monitored. Quantification of the major product gases and tar components will be done off-line. Minor species such as hydrogen sulfide, carbonyl sulfide, and other tar compounds will be monitored if present above the detection limits of the instrument (1-10 ppmv), but will not be quantified. Ammonia, although likely to be present at higher concentrations, is also difficult to determine quantitatively because of interference from other species. Excluded from TMBMS monitoring are carbon monoxide, ethylene and nitrogen. Because these species share the same mass, distinguishing them with a mass spectrometer is not trivial. They are covered by the continuous process gas chromatograph (GC).
2. Determine the effectiveness of the hot gas-conditioning reactor for tar removal, water-gas-shift and perhaps detect changes in hydrogen sulfide and ammonia concentrations. The TMBMS will be used to monitor both sample

points in a sequence to be determined by the experimental design and gasifier operator needs. Typically we will monitor a given process stream for a period of tens of minutes, obtaining a 2-350 amu mass spectrum every 20-30 seconds. This frequency could be increased to every 0.5 second if necessary, with the penalty of filling the data storage capacity rapidly.

Aqueous condensates, which often have been neglected, are measured in the raw gas and product gas streams. Condensate losses between these stations contribute to thermal losses in the system, and high condensate levels entering the turbine can cause problems in the fuel forwarding system.

The fate of organic condensables is a major concern. The small amount of tars that is produced must either be removed by the gas cleanup system or reformed by the gas conditioning catalyst. If they can be reformed they can make a contribution to the heating value of the product gas, and simplify the disposal of scrubber effluent. If removed in the scrubber, they are disposed of by being fed to the fluidized bed combustor vessel of the gasification plant but the residual aqueous effluent must meet environmental standards. This effluent may also contain ammoniacal liquor, dilute acids, and dissolved salts. From a practical standpoint these factors can be critical in deciding the acceptability of a biomass fuelled system.

The TBMS will be used to obtain data that will be compared with data from a smaller scale PDU at the National Renewable Energy Laboratory (NREL) (Ratcliff, et. al., 1998) and ultimately it is hoped that the same apparatus will be used on the pilot plant at Burlington. Data from the latter will necessarily be less comprehensive because of more limited access, but should be sufficient to establish the extent to which laboratory development units can provide valid predictions of full-scale behavior.

Utilization of Data

Data obtained in the course of this work will be used in several ways, including the following:

- To establish correlations between the behavior of the Burlington field demonstration unit, the Battelle process development unit, and the NREL Thermochemical Process Development Unit (TCPDU) (Ratcliff, et. al., 1998), which simulates indirect gasification covering both major and minor constituents in the process streams.
- To assist interactively in process optimization at the Battelle and Burlington sites.
- To contribute to detailed evaluation of the overall process and process steps.

- To provide a quantitative basis for further process improvement.

The ability of TMBMS to continuously follow concentrations of multiple species in real time has proven extremely valuable in verifying stable process operation, detecting transient events or process upsets and diagnosing equipment problems. Figure 2 shows the concentration versus time profile of benzene before, during, and after mechanical problems with the bagasse feed system during gasification tests at the Institute for Gas Technology (Ratcliff, et. al., 1995). The period 8-24 minutes shows how the concentration of benzene (and all other monitored species) varied regularly with time. TMBMS operators determined that this concentration swing corresponded with the periodic operation of the feed lock-hopper system, which resulted in the bagasse being fed in discrete batches. Other on-line instrumentation, which included NDIR gas analyzers, a gas chromatograph, and pressure and temperature monitors, had not detected this instability largely because of inadequate temporal resolution.

Between 24 and 57 minutes bagasse feeding was interrupted as a result of lock-hopper difficulties. Around 57 minutes the feeding problems were resolved and the major product concentrations were roughly equivalent to those prior to the feed interruption, but without the periodic component concentration swings. This was the direct result of a change in feeder operation that maintained a higher inventory of bagasse in the pressurized lock-hopper. The higher feed level apparently stabilized the flow of bagasse on the feed injection screw and isolated the gasifier from the pressure surges associated with the lock-hopper operation.

The ability of the TMBMS to monitor simultaneously on line the values of multiple gas and vapor phase species will enable a much better understanding of the production of compounds such as polycyclic aromatic hydrocarbons, and of their transformation in the catalyst bed. These compounds and the higher hydrocarbons in general, are difficult or impossible to monitor effectively in any other way. The inclusion of water vapor in analyses is also important, as it is a major part of the materials balance. In practical terms, water vapor in the product gas represents a large inert constituent which must be compressed and which can have a major effect on combustion behavior in the turbine because of its effect on flame temperature and (more critically) on flame stability if condensation occurs in the fuel delivery system.

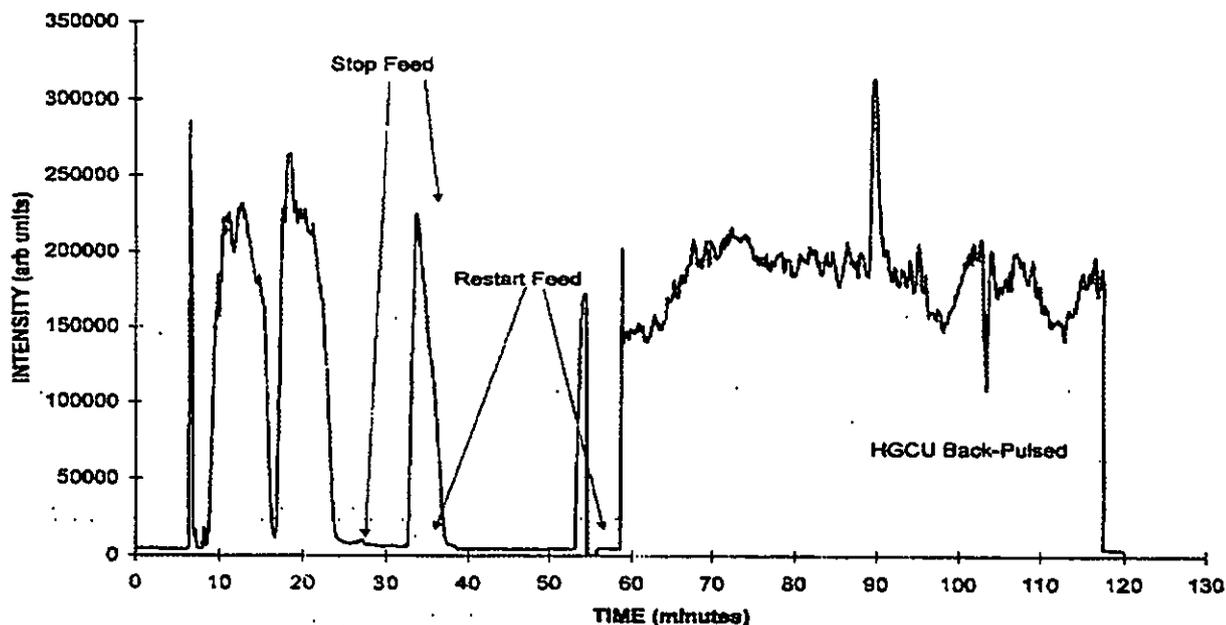


FIGURE 2. BENZENE CONCENTRATION AT HOT GAS FILTER OUTLET.

Inferences for Commercial Applications

Information generated in this program has direct relevance to commercial applications in ways that affect turbine design, system optimization, and permitting issues.

The Solar Spartan turbine will operate in the temperature range 870-500 °C (1600-900 F) at which deposition of alkali compounds may be expected. At the higher temperatures applicable to modern high performance turbines similar conditions may apply in the second or third stages, but the first stages will not be represented directly in this work. However, the retrieval of quantitative data on the composition of solids in the gas stream, and of the deposits themselves, will be valuable in the design of high temperature materials test rigs, which are the accepted vehicles for evaluating turbine hot section materials. Such rigs, incorporating cooled test specimens, can cover a controllable range of gas temperatures, surface temperatures, and thermal gradients, which would be impractical in a full engine test. The Spartan fuel control system is relatively sensitive to fuel quality by virtue of its small physical size. Any condensation or accumulation of tars will produce noticeable effects in control system behavior more rapidly than a larger system. Problems if any, experienced by the Spartan, will have direct relevance to more advanced engines relying essentially on similar control principles.

Moreover, the specific fuel contaminants contributing to such problems, and their origins in the biomass processing system, will be directly traceable. Thus, the problem of ensuring compatibility between all segments of the system, from the raw biomass to the turbine, and ultimately to the system emissions, can be addressed as an entity.

For commercial plant construction and operation, permits must be obtained from local and state regulating boards. The permits required include solid, liquid, and gaseous discharges from the combined cycle plant. The data being developed at Battelle will provide input to each of these permits and allow plant designers to use data specific to the gasification process and the associated gas cleanup and power generation systems.

One of the primary goals of the Battelle program is to provide a gas cleanup system that produces no waste water, thus eliminating a major permitting requirement and providing additional options for waste heat recovery within the gasification plant. Data generated, therefore, must show the fates of organic species such as benzene or polyaromatic hydrocarbons (PAH) as well as any regulated particulates such as heavy metals that might have contaminated the incoming biomass. In the present studies we do not anticipate finding heavy metals, but they can be significant contaminants when recycled materials (construction wastes) are included in the feedstock.

Emission data generated from the full load turbine operation will serve as input to the air permits required for a commercial scale plant operation. The data generated will quantify the effects of removing nitrogen and sulfur containing species from the product gas on NO_x and SO₂ emissions. Such input will allow plant designers to achieve optimal control systems for an integrated plant.

Organic and particulate emissions are also of concern to regulators. Data generated during the testing program will provide combined system specific information on the expected emissions of these pollutants thus simplifying the review of permits by regulators and, therefore, minimizing the time necessary for permit approval.

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