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Used Agricultural Plastic Mulch as a Supplemental Boiler Fuel

- A Report on Combustion Test Results -

Part 1: Streamlined Report

for the

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<u>Note</u>: This report, Part 1, is abbreviated and is written for readers who may not have training or interest in combustion principles or emissions testing and analysis. The complete report describing technical test methods and procedures can be referered to in Part 2: Full Scientific Report.

EXECUTIVE SUMMARY OF COMBUSTION TEST RESULTS

A series of batch-scale stoker combustion tests were performed to evaluate the emissions of:1) criteria pollutants, namely carbon monoxide, sulfur dioxide, and nitrogen oxides; 2) polycyclic aromatic hydrocarbons (PAHs); and 3) dioxin toxic equivalents (TEQ). For emissions reference purposes, baseline combustion tests were run burning coal alone. Additional testing entailed burning the baseline coal blended with plastic fuel nuggets made from discarded agricultural (watermelon) mulch films. Samples were obtained from growers in California, Pennsylvania, and Florida. The plastic nuggets comprised 5% and 10% of the thermal input of the fuel blend.

Regarding criteria pollutants, tests revealed that SO_2 and NO_x emissions using the nugget/coal blends were similar to those when coal alone was burned. CO emissions were very variable, which is a deficiency of batch-type tests.

Regarding PAH and dioxin TEQs, the addition of plastic nuggets to the fuel blend did result in elevated emissions for two of the three plastics used. The emissions from the California plastic tests were similar to those observed when firing only the baseline coal. Complex PAHs and dioxin TEQ emissions were elevated when firing the Pennsylvania and Florida plastics, with the Florida plastic tests exhibiting the highest level of PAHs/dioxin TEQ emissions. The emissions from the Florida plastic tests contained the greatest quantity of the more toxic compounds. Although opinions vary, agreement on two points is generally accepted as 'rules-of-thumb' within the combustion industry: PAH toxicity increases with molecular complexity, and PAH benzo(a)pyrene is considered to be only 20 times less toxic than 1 dioxin TEQ. The PAH concentrations in the Florida emissions should therefore be considered significant.

For future testing, steady-state operation in a large test unit or full-scale system would address the O_2/CO variability and would provide emissions data more comparable to an actual boiler. Pesticide residues on the mulches were not analyzed. The variability in dirt and pesticide contamination prevalent on most waste plastics could be better addressed through utilizing a fluidized bed combustion system rather than a stoker system. A fluidized bed combustion system can handle fuels that exhibit more variability and contain higher ash contents. It is recommend that future testing using plastic nuggets be performed using pilot-scale fluidized bed combustors, or if stoker boilers are to be considered further, a full-scale stoker boiler test should be performed to eliminate the deficiencies of batch-scale testing.

INTRODUCTION

Plasticulture has been an economic salvation for many fruit and vegetable growers worldwide. However, the disposal of these plastics has been difficult for the farmers to manage. Traditional on-farm disposal options such as open burning and on-farm dumping are becoming environmental liabilities. To compound the problem, recycling currently is not an economic management option for dirty plastics, while landfilling the plastic costs more than farmers are willing to pay.

The Pennsylvania State University has developed a process to densify all types of dirty plastics into fuel nuggets. The nugget process will redirect used agricultural plastics from on-farm burning and dumping sites into a new raw material stream for energy recovery when co-fired with coal in community and agricultural boilers. Although the fuel nugget production process works well, the combustion of the nuggets has remained undocumented until this series of laboratory-scale tests was conducted and reported.

Samples of plastic mulch film (low density polyethylene, or LDPE) used in the production of watermelons were received from growers in three states: California, Pennsylvania, and Florida. Growers were asked to follow a specified sampling procedure after the 2001 harvest was complete, which included documentation of all pesticides applied to the watermelon crops and packaging the samples in self-mailer packages. Due to circumstances beyond our control, only one grower supplied the plastic in a timely fashion and listed the pesticides applied during the growing season. Due to insufficient history of usage data, pesticide residues were not analyzed in this investigation.

Research was conducted at Penn State's The Energy Institute using a traveling-grate stoker simulator. The simulator was selected because similar types of combustion units are prevalent in coal-fired community boilers and agricultural operations. Tests were performed firing 100% coal as well as blends of nuggets and coal, with plastics added to provide 5% and 10% of the energy content of the fuel. The flue gas from the stoker simulator was sampled and analyzed, per EPA test protocol, for CO_2, CO, SO_2, NO_x, O_2, polycyclic aromatic hydrocarbons (PAHs), and polychlorinated dibenzodioxins and polychlorinated dibenzofurans (PCDD/Fs) toxic equivalent emissions.

A total of 39 combustion tests were performed. The original test matrix consisted of 21 combustion tests – triplicate tests of the baseline coal, Florida 5%, Florida 10%, California 5%, California 10%, Pennsylvania 5%, and Pennsylvania 10%. However, additional tests were conducted for various reasons, discussed in detail in the results section.

EXPERIMENTAL PROCEDURES AND METHODS USED

Plastic Nugget Maker

The process of making plastic fuel nuggets was developed for waste agricultural plastics, but will also work with plastics found in all sectors of society. Both film and rigid thermoplastics can be accommodated. The process uses a hydraulic cylinder to force plastic items through a heated extrusion die, thereby melting a thin layer of plastic which forms a jacket to seal in plastic pieces and any contaminants. As the extrudate exits the die, a heated cutoff knife melts the end portions to ensure that all pieces of plastic and debris remain encapsulated. The densified plastic cools and becomes a rigid nugget which can be safely stored and easily shipped.

All samples from the growers arrived wet, but were allowed to air dry for three hours prior to being made into fuel nuggets. The California lot included LDPE drip irrigation tubing which had become completely filled with mud, thus the mud became part of the nuggets. The Florida samples were covered with sand, thus the sand became part of the nuggets. Likewise, the Pennsylvania film was contaminated with clay and plant material, which became part of the nuggets. All nuggets were cut into approximately one gram-sized pieces to be of similar size to the coal particles used in the stoker simulator.

Stoker Simulator Test Unit

General Discussion of Coal Combustion on a Traveling Grate

Combustion of coal involves bringing air into contact with the fuel to completely convert all the carbon to CO_2 and hydrogen to H_2O within the residence time available in the combustion chamber. Although this appears to be a simple process, the practical aspects of achieving this objective are far from being simple. In stokers, a variety of physical, chemical and petrographic characteristics govern the combustion behavior. A schematic diagram of a section of a fuel bed on a traveling-grate stoker is shown in Figure 1. The raw fuel, which is usually graded in size, is fed onto the grate and exposed to the ignition source. The bed height usually varies from 4-4.5 inches. The primary source of ignition energy in a large stoker boiler is radiation from the arch and hot combustion chamber walls. Once the top layer of fuel has ignited, heat is transferred to lower layers by radiation and convection, which initiates further combustion. In Penn State's stoker simulator, the ignition source is electrically-heated silicon carbide rods.

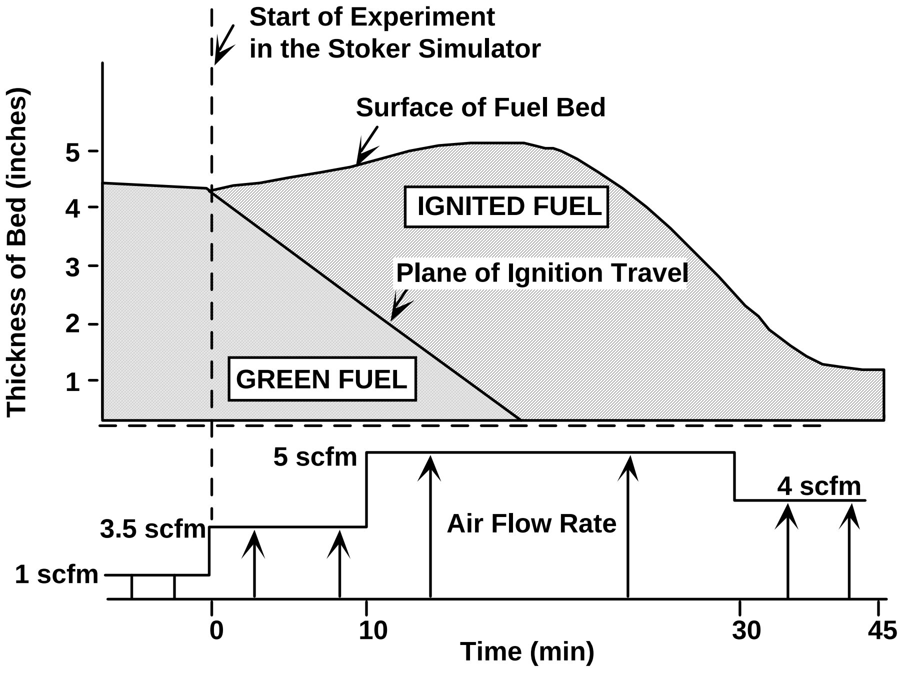

Figure 1. Combustion of fuel in a traveling grate stoker -- section of a fuel bed with time. Scfm is standard cubic feet per minute.

Combustion of coal on a traveling grate can be divided into three zones:

1) Ignition zone -- where ignition occurs initially at the top of the bed and the bed height remains constant. In this zone, the fuel devolatilizes and the volatiles burn above the ignition plane. The plane of ignition travels slowly downwards to the grate;
2) Combustion zone -- where the semi-coke formed after ignition and devolatilization is burned. The semi-coke may swell and resist the flow of air, resulting in an increase in the pressure drop across the fuel bed. The development of plasticity increases the pressure drop, which measures the resistance to the flow of air. Various properties such as the rank of coal, particle size, operating pressure and petrographic characteristics determine thermoplastic behavior. As the ignition proceeds to the grate (with most of the bed being in the burning stage) the air requirement for combustion increases; and
3) Burn-off zone -- where the surface of the semi-coke or coke bed recedes as burning continues, leaving residual ash and clinkers containing some combustibles. The combustion air requirement in this zone is minimal.

Design and Operating Procedure of the Bench-Scale Stoker Simulator

A stoker simulator, which simulates a section of a traveling grate, was used for the testing in this project. Test time is used to replace the distance the grate has traveled with respect to changes in air requirement, bed density, bed height, and residence time.

A schematic diagram of the apparatus is given in Figure 2. The chamber is constructed from a 14 inch steel pipe with 1.25 inches of refractory material to withstand 3,000°F, and 3.5 inches of insulation to minimize heat loss. The coal charge is typically

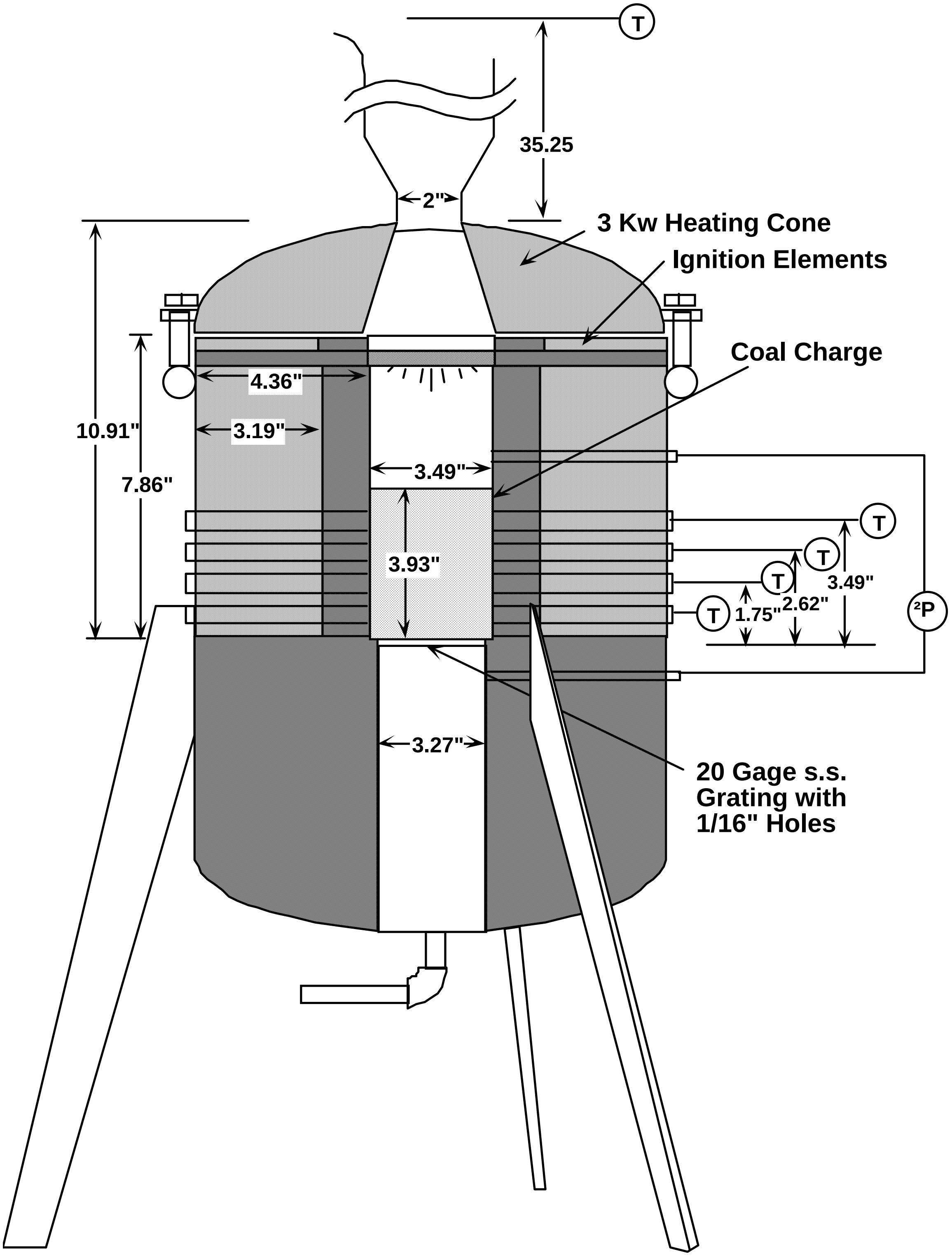

Figure 2. Schematic diagram of the modified stoker simulator.

around 800g (1.76 lbs), which corresponds to a bed depth of about 4.5 inches. The weight of the coal charge corresponds to the mid range area firing density of large-scale utility stokers (8-50 lbs/ft^2).

Combustion air to the stoker simulator is supplied at the bottom of the chamber through a distributor plate. The air flow rate is varied during a test to simulate the variable air flow rate in an operating stoker boiler. During each of the 45 minute tests, the air flow rate for the first 10 minutes is set at 3.5 standard cubic feet per minute (scfm), then changed to 5 scfm for the next 20 minutes and then lowered to 4 scfm for the last 15 minutes. The variation of air flow rate with time is shown in Figure 1. During the peak combustion period at an air flow rate of 5 scfm, the cold air velocity through the grate is approximately 1 ft/s. Although this velocity is lower than the conventional velocity of 1.2 to 1.5 ft/s, it is consistent with the trend of reducing the combustion air flow rate through the grate and increasing the overfire air flow rate to minimize pollutant emissions.

Four 3/8 inch diameter sampling ports are installed at 90° angles to each other at one-inch intervals above the grate. The ports protrude into the coal bed to allow for the measurement of either gas concentration or temperature profile. Type S thermocouples are installed in the reactor, four in the coal bed and one at 14 inches above the top of the coal bed to measure the temperature profile and the exhaust flue gas temperature. The pressure drop across the bed is measured using a pressure transducer to monitor the physical state of the fuel bed during combustion.

A gas sampling port is installed at the simulator outlet to extract flue gas for collection and measurement. A tee was installed for this project and a portion of the flue gas was extracted through an EPA sampling train (described in the next section) while a portion was extracted to a continuous emissions monitoring system (CEM).

A computer was used to collect the temperatures, the gas concentrations, and the pressure drop across the bed.

Continuous Emissions Monitoring Sampling Procedures

The flue gas analyzers used conformed to the following EPA Methods:

Method 3A	Determination of Oxygen and Carbon Dioxide Concentration in Emissions from Stationary Sources (Instrumental Analyzer Procedure);
Method 6C	Determination of Sulfur Dioxide Emissions from Stationary Sources (Instrumental Analyzer Procedure);
Method 7E	Determination of Nitrogen Oxide Emissions from Stationary Sources (Instrumental Analyzer Procedure);
Method 10	Determination of Carbon Monoxide Emissions from Stationary Sources.

Polycyclic Aromatic Hydrocarbons (PAHs) and Polychlorinated dibenzodioxin and Polychlorinated dibenzofuran (PCDD/Fs) Emissions Testing

The Clean Air Act Amendment (CAAA) of 1990 contains provisions that set standards for the allowable emissions of 190 species designated as hazardous air pollutants (HAPS). Many of these HAPS could be emitted from coal-fired electric generating facilities. Polycyclic aromatic hydrocarbons (PAHs) and polychlorinated dibenzodioxins and polychlorinated dibenzofurans (PCDD/Fs) were collected and measured in this project. The techniques used in sample collection and analysis are discussed in the following sections.

Determination of PAH and PCDD/F Concentrations in a Flue Gas Stream

A majority of the 190 species listed in the CAAA including PAHs and PCDD/Fs can be collected by what is generically termed a Modified Method 5 (MM5) sampling train. The train is used to collect samples at desired locations in flue gas streams. The collected flue gas sample can be separated into solid, condensed liquid and gaseous phases. The analytes of interest are extracted from the collected sample, concentrated, then separated and quantified by Gas Chromatography/Mass Spectrometry (GC/MS) and enzyme immunoassay (EIA) techniques.

Analysis and Toxicity of Dioxins and Furans

There are 75 PCDD and 135 PCDF congeners. Of these, 17 are considered to be toxic. Based on a variety of toxicity tests, the EPA has assigned toxic equivalency factors (TEFs) of 1.0 to 0.0001 (relative to 2,3,7,8-tetrachloridibenzo(p)dioxin) to these 17 compounds. In any given sample, the concentrations of each of these compounds can be determined and, when their individual TEFs are factored in, the total toxic equivalency (TEQ) of a sample can be determined. This TEQ is generally considered the data most revealing about a sample’s dioxin-like toxicity.

Method 23 requires use of a high-resolution GC/MS (HRGCMS) for determination of dioxin and furan concentrations. This equipment is quite expensive and not available at The Energy Institute, and analyses performed at commercial laboratories are also quite expensive. TEQ determinations can be made using other methods. The method selected by The Energy Institute is high performance enzyme immunoassay (EIA) colorimetric analysis.

For EIA analyses, a cleaned, extracted sample containing PCDDs and/or PCDFs is added to test tubes coated with a known amount of anti-dioxin antibody EIA. PCDD/Fs are specifically bound to the antibody in proportion to their TEF. A competitor Horseradish Peroxidase (HRP) conjugate is added, binding to the remaining active sites on the antibody. The amount of conjugate bound by the antibody is inversely proportional to the amount of PCDD/Fs present. Color development is inversely proportional to the amount of PCDD/Fs present. Color development is accomplished by adding a solution of chromogenic HRP substrate in hydrogen peroxide. Optical density of the developed color is determined with a spectrophotometer. A series of standards with known TEQ values and optical densities are analyzed, a calibration curve developed, and the TEQ of a sample is calculated.

Methodologies for PAH and Dioxin TEQs

A single 25,500 Btu (British thermal unit) charge of either coal or a coal/plastic nugget blend was placed into the stoker simulator. The fuel was ignited and gas samples containing entrained particulate matter were collected from the stack. Temperature, pressure, and flue gas concentrations were recorded. Samples removed from the stack were analyzed for PAHs and PCDD/Fs. The following EPA test methods were used in collecting and analyzing the samples generated from the stoker simulator for PAHs and dioxin TEQs:

- EPA Method 1A - Sample and Velocity Traverses for Stationary Sources with Small Stacks or Ducts;
- EPA Method 4 – Determination of Moisture Content in Stack Gases;
- EPA Method 23 – Determination of Polychlorinated Dibenzo(p)dioxins and Polychlorinated Dibenzofurans From Stationary Sources;
- EPA Method 3510C – Separatory Funnel Liquid/Liquid Extraction;
- EPA Method 3540C – Soxhlet Extraction;
- EPA Method 3630C – Silica Gel Cleanup;
- EPA Method 8270C – Semivolatile Organic Compounds by Gas Chromatography/Mass Spectrometry (GC/MS); and
- Cape Technologies Method IN-DF1 - High Performance Dioxin/Furan Immunoassay Analysis of PCDD/Fs in Prepared Sample Extracts.

Fuel Composition and Combustion Efficiency

Proximate and ultimate analyses and heating value were determined for the coal and plastic samples. The carbon, hydrogen, sulfur and nitrogen contents of the coal and plastic samples were determined. Heating value of the coal and plastic fuels was determined in an oxygen bomb calorimeter. The ash tracer technique was used to calculate the combustion efficiencies (carbon burnouts) reported in this study.

COMBUSTION AND EMISSIONS TEST RESULTS

Fuel Composition

The composition of the fuels is given in Table 1. The coal used for the testing was a high volatile A coal from the Middle Kittanning seam in Pennsylvania.

Table 1. Fuel composition.

Sample	Parent Coal	California Plastic	Pennsylvania Plastic	Florida Plastic
Proximate Analysis, wt.% (dry basis)				
Moisture	2.0	1.1	0.6	8.0
Volatile Matter	30.6	67.6	85.9	51.6
Fixed Carbon	64.3	2.1	0.5	0.1
Ash	5.1	30.3	13.6	48.3
Ultimate Analysis, wt.% (dry basis)				
Carbon	80.4	55.1	67.2	43.6
Hydrogen	4.8	9.4	11.2	6.6
Nitrogen	1.4	0.3	0.1	0.4
Sulfur	0.7	0.1	0.0	0.2
Oxygen	7.6	4.8	7.9	0.90
Ash	5.1	30.3	13.6	48.3
Chlorine (ppm)	1,538	2,513	2,523	2,775
Higher Heating Value, Btu/lb	13,904	13,011	15,813	11,641

Compositionally, there was much variation among the plastics. This appears to be due to the level of inorganic contaminants (i.e., ash level) in the plastic samples, which ranges from ≈ 14 to 48 wt.%.

The chlorine content of the plastics was greater than that measured in the coal. The chlorine content of the coal was 1,538 ppm as compared to ≈ 2,500 to 2,800 ppm in the plastics.

Final Test Matrix

The final test matrix contained several more tests than originally proposed, i.e., 39 versus 21. Additional tests were performed because:

- One baseline test did not ignite properly;
- A leak was found in the sampling line leading to the continuous emissions monitoring system (CEMs) after several tests were already performed;
- The CO_2 analyzer malfunctioned during a test;
- To obtain additional data to determine repeatability; and
- To obtain additional data to ensure that there was no bias occurring during the testing.

Baseline Tests

The primary PAH compound observed during the baseline testing is naphthalene, which is consistent with other work at Penn State when firing coal in a boiler [1]. The naphthalene concentration varied an order of magnitude from 18 to 161 ng/dscf, which is also consistent with previous work [1]. Naphthalene forms at lower temperatures and its rate of conversion to other compounds is greater than its rate of formation as the temperature increases (e.g., >1,300°C) [1]. As naphthalene conversion increases, more complex PAHs form.

To date, legislation limiting concentrations of PAH emissions from stationary sources has not been enacted. However, under USDOE Contract DE-AC22-93PC93251, [2], flue gas samples were collected prior to an electrostatic precipitator (ESP) in a 108 MW pulverized bituminous coal burning power plant in Niles, Ohio. The naphthalene concentrations in the flue gas stream were found to be 6 ng/dscf. This concentration is approximately 15 times lower than the average concentration found in the baseline stoker tests.

The dioxin TEQ emissions during the baseline tests ranged from 29 to 88 pg/dscf (total of vapor and solid concentrations) and averaged 58 pg/dscf. Although no EPA limits for allowable dioxin TEQ concentrations exist for coal-fired units, the allowable dioxin TEQ limit for medium to large hospital/medical/infectious waste incinerators is 700 pg/dscf [3]. Dioxin TEQ at the Niles Station power plant was found to be approximately 275 fg/dscf. The dioxin TEQ concentrations for the stoker baseline tests fall between these values.

Tests Using California Plastic

The CEM emissions are comparable with the baseline coal emissions. Likewise the range of the PAH/dioxin TEQ emissions, 26 to 86 pg/dscf (total solid and vapor), is similar to the baseline tests. However, the tests conducted with 10% plastics tended to have slightly lower PAH and dioxin TEQ emissions. Again, these concentrations fall between the concentrations allowed for incinerators and what was found at the Niles Station utility boiler.

Tests Using Pennsylvania Plastic

The CEM emissions are comparable to the baseline coal emissions; however, the complex PAH and dioxin TEQ emissions are elevated. The tests with more plastic (i.e., 10% by thermal input) tended to have higher levels of complex PAHs and dioxin TEQ emissions. From Niles Station, concentrations of phenanthrene were found to be 10 ng/dscf, fluoranthene was 3 ng/dscf, and pyrene was 1 ng/dscf.

Tests Using Florida Plastic

As with the Pennsylvania and California plastic tests, the CEM data is comparable to the baseline tests. The PAH and dioxin TEQ emissions however, were significantly elevated and contained many complex PAHs. These results were the worst of all of the testing and are sufficiently high to cause concern. The Florida plastic contained the highest level of chlorine and was the most contaminated (i.e., ≈ 48 wt.% ash) of the three plastics. Testing from Niles Station showed the following concentrations of PAHs: fluorine at 4 ng/dscf, anthracene at 1 ng/dscf, benz(a)anthracene at 1 ng/dscf, benzo(a)pyrene at 600 pg/dscf, and benzo(g,h,i)perylene at 50 pg/dscf.

Concluding Statements

A series of batch-scale stoker combustion tests were performed to evaluate the emissions of criteria pollutants (carbon monoxide, sulfur dioxide, and nitrogen oxides), polycyclic aromatic hydrocarbons (PAHs), and dioxin toxic equivalents (TEQ). Tests were conducted using a baseline coal and blends of the baseline coal with three plastic mulches (obtained from California, Pennsylvania, and Florida) with the plastic nuggets comprising 5 and 10% of the thermal input of the fuel blend.

The SO_2 and NO_x emissions from the tests using the nugget/coal blends were similar to those firing only coal. CO emissions were very variable which is a deficiency of batch-type tests. In normal boiler operation, the oxygen content is monitored and the quantity of air used for combustion is adjusted to maintain a constant O_2 level, e.g., 4%, in the flue gas. However, in the batch scale tests, the air flow rate was maintained constant for all tests to ensure uniformity between tests, especially because of the PAH/dioxin TEQ sampling. Hence, variations in O_2, which adversely affects CO as the O_2 level drops very low, are experienced due to varying combustion efficiencies, clinkering in the bed, and, in the case of coal/nugget blends, possibly different combustion profiles.

The addition of plastic nuggets to the fuel blend did result in elevated PAH/dioxin TEQ emissions using two of the three plastics. The emissions from the California plastic tests were similar to those observed when firing only the baseline coal. Complex PAHs and dioxin TEQ emissions were elevated when firing the Pennsylvania and Florida plastics with the Florida plastic tests exhibiting the highest level of PAHs/dioxin TEQ emissions. The emissions from the Florida plastic tests contained the greatest quantity of the more toxic compounds. Although opinions vary widely, agreement on two points is generally accepted as ‘rules-of-thumb’: PAH toxicity increases with molecular complexity and benzo(a)pyrene is considered to be only 20 times less toxic than 1 dioxin TEQ. The PAH concentrations in the Florida emissions should therefore be considered significant.

Steady-state operation in a larger test unit or full-scale system would address the O_2/CO variability and would provide emissions data more comparable to an actual boiler. The variability in plastic contamination could be better addressed through utilizing the plastics in a fluidized bed combustion (FBC) system rather than a stoker system since an FBC can handle fuels that exhibit more variability and contain high ash contents. It is recommend that future testing using plastic nuggets be performed using pilot-scale FBCs or, if stoker boilers are to be considered further, a full-scale stoker boiler test should be performed to eliminate the deficiencies of batch-scale testing.

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