Emissions from a domestic two-stage wood-fired hydronic heater: Effects of non-homogeneous fuel decomposition

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Abstract
Accurate knowledge of the combustion gas composition is necessary for practical combustion systems where emissions reduction is a major concern. Industries use a variety of sensor technologies, many of which can be expensive. In industries that use plant biomass for heat production, a constant fuel composition is often assumed for estimation of unmeasured exhaust gas species and for the calculation of thermal efficiency. To address the fact that biomass decomposes non-homogeneously, a new analysis is developed where a 2:1 ratio of hydrogen to oxygen atoms in the fuel is assumed, thereby relaxing the constant fuel composition constraint. This analysis provides a reliable and accurate estimate of exhaust $H_2O$ and $CO_2$ given commonly monitored air and fuel rates along with $O_2$, $CO$, and $NO$ measurements. Validation of the analysis is investigated using tunable diode laser absorption spectroscopy (TDLAS) measurements of the flue gas. Results show good agreement between the new analysis and TDLAS measurements, verifying the validity of the approach. The new analysis also provides time accurate elemental species information for interpreting the non-homogeneous fuel decomposition.

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1. Introduction

The environmental advantage of using biomass over fossil fuel has been widely discussed for power generation and heating [1–3]. Many developing nations rely exclusively on biomass for energy production, as well as rural areas in northern regions of the United States where wood fired heaters are an economically attractive alternative. However, one of the prevailing challenges using these systems is emissions. For the northeastern US, emissions from wood fired heaters are one of the leading sources of reduced air quality. To provide stricter guidelines on the certification of these systems, the Environmental Protection Agency (EPA) has recently defined specific new regulations for wood burning appliances including wood stoves, masonry heaters, pellet-burning stoves and hydronic heaters [4]. Many studies focus on the characterization of emissions from specific biomass appliances, stoves, boilers, and large scale (> 1 MW) power plants [5,6]. Reduction in harmful pollutants resulting from the combustion of cellulose based biomass materials is the primary goal of many government agencies and research developments.

Certification of these devices require an accurate representation of emissions species concentrations. Measurements of both $CO_2$ and $H_2O$ are typically required for emissions-based efficiency calculations [7]. For many independent testing facilities and manufacturers, direct measurement of gas species can be cost prohibitive. Often, low cost gas sampling analyzers (Testo, Land, Wohler, etc.) are used to measure select harmful pollutants ($CO$ and $NO$) directly, along with the oxygen sensors to estimate $CO_2$ concentrations based on an assumed constant fuel composition. In conjunction with the $CO_2$ estimation, $H_2O$ is often approximated using a global reaction mechanism [4,7–16]. It is therefore subject to interpretation and inaccurate for some conditions. While non-homogeneous biomass decomposition has been extensively studied [8,14,17–21], its application in the biomass community has been relatively unexplored.

The facility and analysis for a constant fuel formulation (CFF) was developed in Ref. [15]. This study builds on reference [15] by presenting a novel variable fuel formulation (VFF) to accurately calculate the non-homogeneous decomposition of the solid fuel and major exhaust gas species ($CO_2$ and $H_2O$) using measurements commonly made in the biomass industry. Exhaust gas concentrations ($O_2$, $CO$, and $NO$) and mass flow (fuel and air) measurements are used in an atom balance, assuming a 2:1 ratio of hydrogen to oxygen atoms in the fuel. Validation of the analysis is demonstrated...
through non-intrusive tunable diode laser absorption spectroscopy (TDLAS) measurements of CO$_2$ and H$_2$O in the flue. CO$_2$ and H$_2$O concentrations have been previously measured using TDLAS in combustion environments [22,23]. The results from the TDLAS measurements are based on first-principles quantum spectroscopy theory, where single molecular absorption transitions are monitored to extract information regarding concentrations and temperatures of the absorbing gas species.

The novelty of the following analysis is to provide a method for calculating in real-time the changing fuel composition and major flue emissions (CO$_2$ and H$_2$O). The direct measurement of important major combustion species CO$_2$ and H$_2$O at elevated temperatures can often be cost-prohibitive. This analysis provides an accurate approach for their calculation using low-cost, routine measurements. This study also demonstrates that real-time fuel characterization for practical biomass combustion systems can improve efficiency calculations over those using a constant fuel assumption.

The rest of this study is organized as follows. Section 2 will discuss the experimental setup used in the analysis. Section 3 will detail the analysis used to calculate the exhaust species and non-homogeneous elemental fuel composition. Section 4 will discuss the results of the CO$_2$ and H$_2$O validation with TDLAS. Finally, Section 5 will discuss the major conclusions and outline the novelty of this work.

## 2. Facility and baseline measurements

### 2.1. Hydronic heater experimental setup

The test facility previously discussed in Ref. [15] is used in this study where an Econoburn (Brocton, NY) EBW-200 wood-fired hydronic heater (WFHH) is instrumented. The setup consists of a primary and secondary water circulation loops, and a heat exchanger to expel heat. Fig. 1a shows the boiler setup, where the load loop contains a 300,000 BTU/h (87.9 kW) counterflow heat exchanger to transfer heat from the boiler to a cold thermal sink. The heat rate is monitored by recording the water temperatures and flow rates, providing a detailed trend of the supplied heat load.

#### 2.1.1. Measurements of fuel and airflow

Instantaneous fuel mass burn rate is measured according to the method presented in Richter et al. 2016 [15] (see Fig. 1b). The real time fuel burn rate monitor (RTFBRM) is designed to directly measure fuel mass loss in the upper chamber as a function of time. The assembly consists of a basket suspended by two rods which run through the top of the boiler and are sealed using Swagelock fittings. Outside of the boiler, the rods are connected to a horizontal upper cross member. The upper cross member rests on a specially designed piezoelectric-based compression load cell (Stellar Technology) which is thermally insulated from the boiler to avoid any biases induced by thermal gradients. Contact friction is minimized by using high-temperature carbon laced rope around the rods to ensure an accurate instantaneous mass measurement using LabVIEW data acquisition systems (National Instruments) at a frequency of 2 Hz. Fig. 2a shows the raw mass data and Fig. 2b demonstrates the differentiated mass loss rate with time. These measurements are used in the emission analysis discussed in section 3.

To minimize batch-to-batch variability associated with boiler operation, BIOBLOCKS®, manufactured by Summit Wood Industries, are used as the primary fuel source. The blocks are made from 100% hardwood chips (composed primarily of red oak) and contain an average 8.3% dry-basis by mass water moisture. The exact moisture content and density of the blocks are measured in accordance with ISO standard 3130 resulting in a density of 1.15 g/cm$^3$. A full load boiler run requires 32 blocks (≈62 lbs, 28 kg) to conform to current testing standards. A standard loading configuration has been adopted to produce a repeatable burn [15].

Blower air flow rates are measured using a Bosch HFM-7 mass airflow meter operating under hot film anemometry methods. Direct calibration of the Bosch meter was made with an ASME standard venturi flow meter across a wide range of flow rates to correlate output voltage to measured flow rate. Fig. 2c shows the measured air flow rate as a function of time that will be used in the emissions analysis. The sharp decrease in air flow rate seen in Fig. 2c at approximately 0.35 h is caused by the appliance damper being pulled, effectively increasing the blockage through the appliance by forcing the incoming air into the secondary and tertiary chambers before exiting. The gradual rise in flow rate starting at approximately 3 h is due to the gradual decrease in blockage as the fuel decomposes.

### 2.1.2. Measurements of CO, NO, O$_2$, and gas temperatures

Emissions measurements are recorded using a Testo 330-2LL gas analysis meter. The analyzer is capable of taking experimental measurements of the O$_2$, CO, NO, pressure, and temperature from the flue gas stream. The O$_2$ measurement has a range of 0 to 21% with a resolution of 0.1%. Oxygen concentration is detected via an electrochemical fuel cell with a permeable layer of electrodes that allow the flue oxygen molecules and reference air molecules to transfer ions, creating a voltage potential across the electrodes that...
can be interpreted as an O$_2$ concentration within the flue gases. Similarly, the CO concentration is also measured via an electrochemical fuel cell, but differs in that O$_2$ molecules that permeate the electrodes are used to react with the CO on the opposite electrode to form CO$_2$. This chemical reaction results in the consumption of O$_2$ on the reference side of the fuel cell and is used in conjunction with the electrode at the reaction location to obtain a voltage potential that can ultimately be interpreted as a concentration of CO. This results in a CO measurement in parts per million (ppm) from 0 – 30,000 at a resolution of 1 ppm. Operating on a similar principal as the CO sensor, an electrochemical fuel cell is used to determine the concentration of NO in a range of 0 – 3,000 ppm with a 1 ppm resolution. The CO$_2$ recorded by the Testo analyzer is estimated from the O$_2$ measurement and limited by the maximum theoretical value of CO$_2$ based on the fuel composition at the stoichiometric limit. The Testo sampling probe, placed in the exhaust gas stream, includes a thermocouple and inlet for flue gas sampling. To correct measurements for water condensation in the sampling line, the correction procedure of Richter et al. [24] is used, requiring a separate measurement of O$_2$ using a Bosch LSU 4.9 ZrO$_2$ wide-band oxygen/lambda sensor. Ambient moisture content and temperature are measured in real-time with an Omega HX92B series humidity sensor and K-type thermocouple. Fig. 3 shows mean measured O$_2$, CO, and NO with data spread from three runs. The mean gas temperatures measured using a bayonet thermocouple mounted in the primary chamber and Testo analyzer in the flue are shown in Fig. 4.

### 3. Emissions analysis

A variable fuel formulation is developed to determine the flue gas composition given the direct measurements of fuel and air flow rate, CO, NO, and O$_2$. In this approach, the wood fuel is assumed to have a variable composition, with the defining characteristic that the hydrogen and oxygen are released at a constant 2:1 ratio [25]. To include fuel nitrogen and reduce unknowns in the analysis, NO emissions are assumed to be derived entirely from the fuel, consistent with earlier studies [6,19,26,27]. This assumption is supported by the recent results of Rabacal et al. 2013 [6] where measured NO emissions are linearly correlated with fuel nitrogen.
Fig. 2. Comparison of (a) mass, (b) burn rate, and (c) air flow rate data for several runs.

Fig. 3. Comparison of (a) $O_2$ mole fraction, (b) $CO$ mole fraction, and (c) $NO$ mole fraction data.
While there are various products of incomplete combustion, CO and NO are the only ones considered in this analysis since they are the only products that can be measured using the Testo gas analyzer. The sensitivity of the NO was investigated and shown to produce a negligible difference in the results of the following analysis. Other products of incomplete combustion that are on the same order of magnitude as NO are also assumed negligible. Additionally, since CO is an intermediate species of CO₂ production, it is among the more substantial of the minor species and is often used as a direct indicator of combustion performance [12,13,28,29]. The overall one-step reaction for combustion is assumed to have the following form, as:

\[ C_1H_{20}O_n + a(O_2 + 3.76N_2 + \gamma H_2O) \rightarrow cH_2O + d CO + e CO_2 + 3.76a N_2 + g O_2 + h NO \]  

(1)

On the left hand side of the reaction, the unknown wood fuel composition is described as \( C_1H_{20}O_n N_2 \), with the moisture content of the fuel included as part of the composition. The moisture coefficient \( \gamma \) describes the ratio of mole fractions between oxygen and water in humid air, and is calculated from ambient humidity and temperature measurements, leaving three unknowns (\( b, h, \) and \( a \)). On the right hand side of the reaction, the remaining species coefficients \( c, d, e, \) and \( g \), and the total moles, \( N_{\text{tot}} \), leave a total of eight unknowns. Three constraints are provided by measured values of \( CO, NO, \) and \( O_2 \) expressed as \( X_{CO} = d/N_{\text{tot}}, \) \( X_{NO} = h/N_{\text{tot}} \) and \( X_{O2} = g/N_{\text{tot}} \), respectively. Three more constraints come from atomic balances for \( C, H, \) and \( O \) (\( N \) is dependent on the \( NO \) measurement) along with the condition that the mole fractions sum to unity. Closure of the system of equations is obtained from measurements of fuel mass loss and air flow rate,

\[
\frac{\dot{m}_f}{\dot{m}_a} = \frac{MW_C + 2bMW_H + bMW_O + hMW_N}{a(MW_{O2} + 3.76MW_{N2} + \gamma MW_{H2O})}
\]  

(2)

In total, eight equations are solved and are expressed in matrix form as,

\[
\begin{bmatrix}
0 & 0 & 0 & 0 & -1 & 0 & 0 & X_{CO} \\
0 & 0 & 0 & 0 & 0 & -1 & X_{O2} & h \\
0 & -1 & 0 & 0 & 0 & 0 & X_{NO} & a \\
0 & 1 & 3.76 & 1 & 1 & 1 & 1 & c \\
0 & 0 & 0 & 1 & 1 & 1 & 0 & d \\
2 & 0 & Z_\gamma & -2 & 0 & 0 & 0 & e \\
1 & -1 & (2 + \gamma) & -1 & -1 & -2 & -2 & 0 \\
\end{bmatrix}
\begin{bmatrix}
\Omega - MW_N \\
\Psi \\
0 \\
0 \\
0 \\
0 \\
0 \\
0 \\
\end{bmatrix}
= \begin{bmatrix}
b \\
da \\
c \\
d \\
e \\
0 \\
0 \\
191
\end{bmatrix}
\]  

(3)

where the quantities \( \Psi \) and \( \Omega \) are defined as,

\[
\Psi = \frac{\dot{m}_f}{\dot{m}_a} (MW_{O2} + 3.76MW_{N2} + \gamma MW_{H2O})
\]  

(4)

\[
\Omega = (-2MW_H - MW_O)
\]  

(5)

The system of equations are solved for each sample measurement using Gaussian elimination. The resulting analysis provides real-time \( C, H, O, \) and \( N \) atomic ratios for the non-homogeneously decomposing fuel, and emissions mole fraction of species \( CO_2, H_2O, CO, NO, O_2 \) and \( N_2 \) in the flue exhaust.

3.1. Experimental validation: TDLAS measurements of \( CO_2 \) and \( H_2O \)

Tunable diode laser absorption spectroscopy is used to directly measure the concentrations of \( CO_2 \) and \( H_2O \) of the exhaust gas in the flue in order to validate the new analysis. Accurate measurements of mole fractions are obtained because the signal is a result of individual ro-vibrational molecular absorption transitions, and the lineshapes of the gas absorption features are well defined. A schematic of the diagnostic setup is shown in Fig. 5.

The infrared laser diode is a distributed feedback quantum cascade laser constructed by Nanoplas specifically for emission at 2.715 \( \mu \)m, packaged in a TO-5 heat sink housing with a collimation lens on the front. A Lightwave ILX LDC-3721 laser diode driver and thermoelectric cooler (LDD-TEC) is used to control the laser. The diode emits at 2.715 \( \mu \)m when the temperature is 37 \( ^\circ \)C and the current is 155 mA (output power of 5.4 MW). To scan through a wavenumber range, the output current is modulated using a Tektronix AFG3051C arbitrary function generator, with a frequency of 20 Hz. The function generator output is connected to the LDD-TEC input, and modulates the output current of the LDD to the laser. The small, controlled current modulation scans the laser from a low to high wavelength. The wavenumber scan range used for these measurements is approximately 3683.5 \( cm^{-1} \) to 3686.5 \( cm^{-1} \). This range is used to capture both \( CO_2 \) and \( H_2O \) peaks in a single ramp.

Due to incomplete combustion, soot is also present in the exhaust gases, and can accumulate on the \( CaF_2 \) windows. To mitigate this effect, the windows are bathed with \( N_2 \) throughout the duration of a run. To eliminate absorption in the laboratory air, the laser pitch and laser catch sides are housed in acrylic boxes that are purged with \( N_2 \).

Every ten seconds, the system collects four ramps with 10,000 data points (2500 points per ramp). Running at a frequency of 20 Hz, four ramps are scanned over a period of 0.2 s, which is faster than the transients occurring in the boiler. Three detectors are connected to a data acquisition system (National Instruments). The first detector captures the signal through a solid germanium etalon with a free spectral range of 0.73 GHz, which is used to convert between arbitrary time and wavenumber. The second detector captures the absorption signal through the flue gas. The third detector captures a baseline ramp, which is \( N_2 \) purged over its entire line-of-sight such that there is no absorption. This line-of-sight
passes through the baseline tee, which extends from window to window in the flue, and is pumped with N₂ throughout the run.

The data from the three detectors is used to convert the raw signal (arbitrary voltage versus arbitrary time) into the final transmission versus wavenumber signal, which is compared to simulations obtained using a line-by-line code. The path length through the flue gas is known precisely, and the temperature of the gas is known from a thermocouple located just upstream of the measurement. Simulations are performed for various combinations of CO₂ and H₂O mole fractions at the given temperature, and a Chi-square error surface is constructed from which the minimum gives the best-fit to the boiler data. The error surface monotonically decreases in all directions, such that the minimum found from the simulations is assumed to be a global minimum. The error surface, and corresponding raw data and best-fit simulation data can be seen in Fig. 6.

4. Results

A comparison of TDLAS measurements versus emissions estimates of CO₂ (assuming either the VFF or CFF) is shown in Fig. 7. The data bars represent variation in the measurements over three runs. Comparison of the VFF against TDLAS measurements agree well with a mean error of approximately 6%. The CFF formulation, however, under predicts by a mean error of approximately 23%. These differences are due to the preferential release of H₂O early in time which changes the effective composition of C, H, O, and N in the wood. The VFF accounts for these changes, while the CFF inherently does not. The location of the peak CO₂ range (Fig. 7) corresponds approximately to the minimum CO (Fig. 3b). Within this period between 1 and 2.5 h, the excess air ratio is at its minimum (approximately 3), and together with higher gas temperatures (Fig. 4) creates favorable oxidation conditions, resulting in an increase in CO₂ and decrease in CO [30].

The early release of H₂O can be seen in Fig. 8 where a comparison of the calculated VFF and CFF against TDLAS measurements are presented. Excellent agreement is observed between the TDLAS measurements and the VFF with an approximate mean error of 14%. However due to the uniform fuel assumption of the CFF, H₂O is underpredicted at early times and overpredicted during the charcoal oxidation stage, resulting in a mean error of approximately 64%. This direct comparison to TDLAS demonstrates the clearly erroneous assumption of a constant fuel approach, and shows that the VFF provides a more accurate physical description.

4.1. Time history: elemental fuel composition

Red oak has an approximate composition of C₁₁H₁₇O₇N₀.₇₂N₀.₀₀₁ [31], and with the 8.3% dry basis unbound water, is approximately C₁₁H₁₇.₉₄O₀.₆₂N₀.₀₀₁. Since no constraint in the emission analysis has been placed on the exact fuel composition and moisture content, the elemental composition of the fuel can be determined as a function of time. Fig. 9 shows the ratios H/C, O/C, and N/C as a function of time. Distinct stages of drying, pyrolysis/combustion and charcoal oxidation are identified in Fig. 9. In the drying stage, the majority of unbound fuel moisture has been driven from the fuel, resulting in an increase in effective H and O atomic ratios in the fuel. This is further supported by integrating the fuel based H₂O
where approximately 2.2 kg (amount of free $H_2O$ contained in 28 kg of 8.3% moist fuel) has been released at approximately 0.25 h, corresponding with the end of the drying stage. The second stage consists of a combination of virgin fuel pyrolysis and combustion resulting in a linear decrease in $H$ and $O$ with time until the fuel $H$ has been oxidized. Stage three, starting at approximately 2.75 h, is characterized by the full consumption of $H$ and $O$ leading to the charcoal oxidation stage. The nitrogen slowly increases as the remaining charcoal is further depleted.

4.2. Time history: heating value

The time dependent fuel composition can be used to estimate the higher heating value (HHV) of the fuel [20].

$$HHV = \frac{33.5(CC) + 142.3(HC) - 15.4(OC) - 14.5(NC)}{10^2}$$

Equation (6) represents the (HHV) [MJ/kg] for biomass fuel given a specified elemental mass percentage for carbon (CC), hydrogen (HC), oxygen (OC), and nitrogen (NC) [20]. Fig. 10 presents the HHV for the variable fuel composition with comparison to reference values for red oak (with 8.3% moisture content) and coal. The images in Fig. 10 show the progression of the burn at early and late times. As the virgin fuel dries early in time, it undergoes pyrolysis and forms charcoal, and the HHV is reduced (close to the constant value of 17.5 MJ/kg for red oak). The later burning is characterized primarily by charcoal oxidation, where the HHV approaches that of coal (32.3 MJ/kg). The three modes of biomass are shown in Fig. 6.
conversion (drying, pyrolysis and char oxidation) are widely known \cite{6, 20, 32, 34} and are confirmed in these results.

4.3. Time history: thermal efficiency

Thermal efficiency is often expressed in terms of total (integrated) heat delivered ($\Delta Q_{\text{del}}$) divided by the total caloric chemical energy available ($\Delta Q_{\text{comb}}$) of the fuel used, i.e., \( \eta_{\text{th}} = \Delta Q_{\text{del}} / \Delta Q_{\text{comb}} \), where a single efficiency is reported per fuel charge \cite{4}. While this definition provides an estimate of the average efficiency of an appliance, it is limited since important transient information is lost. The missing time dependent information is useful for understanding the boiler dynamics to improve function and design. An instantaneous thermal efficiency ($\eta_{\text{th}}$) is therefore desirable. The practical calculation of it is difficult, since previously the time dependent caloric value of the wood was unknown. Using the results from Fig. 10, $\eta_{\text{th}}$ can now be computed as:

\[
\eta_{\text{th}} = \frac{Q_{\text{del}} + Q_{\text{stor}}}{Q_{\text{comb}}},
\]

where $Q_{\text{comb}} = (\text{MJ/HHV})$ is the heat produced from combustion. The term $Q_{\text{stor}} = C_w m_w (T_{w,\text{out}} - T_{w,\text{in}})$ represents the instantaneous heat transferred to the boiler and inferred from the measured mean water and wall temperatures. The term $Q_{\text{del}} = C_w m_w (T_{w,\text{out}} - T_{w,\text{in}})$ is the delivered heat to the thermal load (heat exchanger) which is experimentally determined from measurements of the water temperature and flow rate going in ($T_{w,\text{in}}$) and out ($T_{w,\text{out}}$) of the boiler. The inverse relationship between the fuel heat rate and the efficiency results in higher efficiency with lower heat rate. This is due to the time lag associated with heat transfer through the boiler. Fig. 11 shows $Q_{\text{del}}$, $Q_{\text{stor}}$, $Q_{\text{combVFF}}$, and $Q_{\text{combCFF}}$ versus time for a typical boiler run. It can be seen that $Q_{\text{del}}$ and $Q_{\text{stor}}$ are inversely proportional, indicating the heat rate delivered from the water is coming from the boiler heat capacitance, rather than from the heat generated from combustion. Compared to the CFF description, the heat release from the VFF is comparatively lower in the beginning and higher at the end, which is consistent with the individual fuel descriptions for red oak and coal. Fig. 12 shows a comparison of the VFF and CFF instantaneous efficiency as a function of time. The efficiency using the CFF heating value (Fig. 11, blue dashed line) results in non-physical values greater than unity. It is only when the heating value derived from the VFF (Fig. 11, red solid line) is used that physically realistic efficiencies are achieved. Integrated thermal efficiencies ($\eta_{\text{hCFF}} = (\Delta Q_{\text{del}} + \Delta Q_{\text{stor}}) / \Delta Q_{\text{comb}}$) calculated with CFF and VFF are comparable at approximately $\eta_{\text{hCFF}} = 50\%$ and $\eta_{\text{hVFF}} = 54\%$, respectively.

5. Conclusions

An experimental facility was modified for further experimental diagnostics, including measurements of fuel and air flow rates and
a TDLAS setup at the flue [15]. Emissions and flow rate measurements are used in the new analysis and validated using direct, non-intrusive, first principles-based TDLAS measurements of CO₂ and H₂O. The analysis provides estimates of major exhaust species and changes in elemental fuel composition using emissions measurements of CO, NO, and O₂ and flow measurements of fuel and air. Good agreement was shown in the validation, comparing predictions of CO₂ and H₂O to TDLAS measurements.

This new analysis provides a clear description of the various stages in fuel decomposition for batch-run wood burning systems. The changing elemental composition of the fuel is calculated without direct knowledge of the exact wood type or unbound moisture content. Three distinct burn stages are identified consisting of drying, pyrolysis/combustion and charcoal oxidation. Using the varying composition, a time dependent HHV is estimated and shown to vary from 10 MJ/kg early in the heating process to 33 MJ/kg. These stages are consistent with the observed burning modes. Lastly, the time dependent HHV is used to determine a time dependent thermal efficiency which is useful for understanding the instantaneous performance of the boiler.

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