The behavior of secondary pyrolysis products in oxy-coal flames

P. Tóth[*,1], M. Aczél[1], T. Draper[2], E. G. Eddings[2]

[1]Department of Combustion and Thermal Energy, University of Miskolc, Hungary
[2]Department of Chemical Engineering, University of Utah, UT, US

Abstract
Advanced, imaging-based optical diagnostics (particle imaging velocimetry, infrared radiation mapping and high-speed shadowgraphy) were used to study the formation and behavior of condensed secondary products in a laboratory-scale pulverized-coal flame. The system allowed for the quantitative description of gas velocities, particle velocities, particle sizes and shapes, particle concentrations and flame temperature. A qualitative assessment of CO_2 concentrations was also carried out. It was found that oxy-firing significantly alters the behavior of secondary products – replacing N_2 with CO_2 in the oxidant mixture suppressed the formation of the condensed tar phase in the studied flame. We aim to identify the driving force behind tar formation and explain why and how oxy-firing changes the distribution of secondary pyrolysis products in a laboratory-scale pulverized-coal flame.

Introduction

Oxy-firing is a potential solution for making pulverized-coal combustion sustainable. In oxy-coal combustion, instead of air, O_2 is used as an oxidant. Pure O_2 is diluted with recirculated flue gas (mostly CO_2) in order to control flame temperature. The main benefit of oxy-coal combustion is the high CO_2 concentration of the flue gas, which makes the capture and reuse of CO_2 possible. Oxy-coal combustion systems are typically built by retrofitting existing, air-fired systems [1].

It is well-known that switching the oxidizer from air to a CO_2/O_2 mixture alters the thermal and aerodynamic behavior of a pulverized-coal flame. Many studies reported altered temperature, radiation, velocity and species profiles when operating a burner in oxy-combustion mode [1]. Another well-known characteristic of oxy-coal firing is the altered stability of the produced flames in terms of flame detachment [2]. The possible cause of the de-stabilization is the ignition behavior of individual coal particles that is changed by replacing the air atmosphere with a CO_2/O_2 mixture.

Several studies focused on the effects of oxy-coal firing on particle ignition from a single particle combustion point of view [3-5]. A study by Galletti et al. found that replacing air with an atmosphere typical in oxy-fuel combustion increases the ignition delay of individual coal particles [3]. Similar results were reported earlier by Shaddix and Molina [6]. Marek and Swiatkowski found that bituminous coal particles burn at lower temperatures in oxy-firing mode than in air firing [4]. The temperature differences up to 70 K were explained by the higher heat capacity of the oxy-atmosphere than that of air. The results of Riaza et al. were in agreement with the lower temperatures observed by Marek and Swiatkowski. Riaza et al. made further observations regarding the ignition temperature of particles (which was increased by oxy-firing), the effect of O2 concentration in the oxy-firing atmosphere (higher O2 concentrations decreased ignition temperature and increased burning temperature by up to 320 K) and the burning mode (bituminous coal particles burned in two-mode combustion, meaning that the volatiles and char burned in distinctly different modes) [5].

Kim et al. used high-speed visible imaging to study the burning modes of particles of different coal types. They reported that among the studied types of coal, two bituminous coal types burned in distinct two-mode combustion, where the volatiles were ejected from the particles and then formed condensates that surrounded the particle. Kim et al. referred to this mode, apparently specific to the studied bituminous coals, as tar-tail combustion [7].

The so-called tar-tails were observed very early by Choi, who used pulsed light imaging to directly observe volatile cloud formation in pulverized-coal flames. In his 1990 paper, Choi proposed a simple mechanism for tar-tail formation – his tests were also carried out by using a bituminous coal type [8]. The mechanism proposed by Choi is summarized by the following figure.

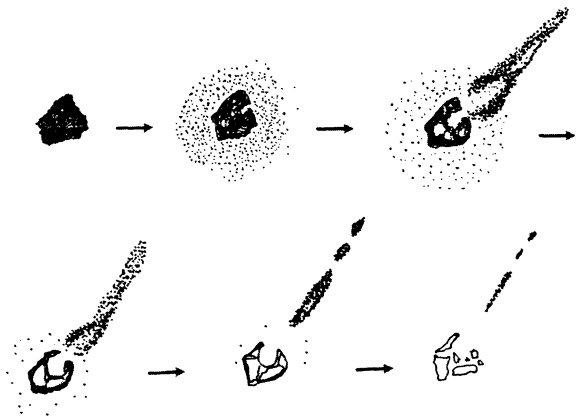

Figure 1: Conceptual diagram of tar-tail combustion: particle heat-up, devolatilization, ejection of volatile jet, condensation, tail breakup and finally char oxidation [8].

* Corresponding author: toth.pal@uni-miskolc.hu

In this work we study the effect of oxy-firing on secondary pyrolysis products (tar and soot trails) formed during the combustion of bituminous coal particles. The concentration and spatial distribution of these secondary products in a pulverized-coal flame are of importance, as they affect the radiation profile of a flame.

Materials and methods

The experimental apparatus consisted of a laboratory-scale pulverized-coal burner and a complex optical diagnostics system. In this section we give the details of both systems.

The burner

Coal was fed into the tube-in-a-tube type pulverized coal burner shown in Figure 2 by a coal feeder unit. This unit consisted of a quartz tube that was slowly raised into a stainless steel tube by an electric motor. Inside the stainless steel tube, another smaller stainless steel tube extended into the quartz tube. Carrier gas (CO_2) was injected into the annulus between the two steel tubes. The gas turned inside the quartz tube and carried coal into the smaller steel tube, which was connected to the central port of the burner. The motor speed and carrier gas flow rate were adjustable – the coal feed rate was precisely set to 0.1 g/min.

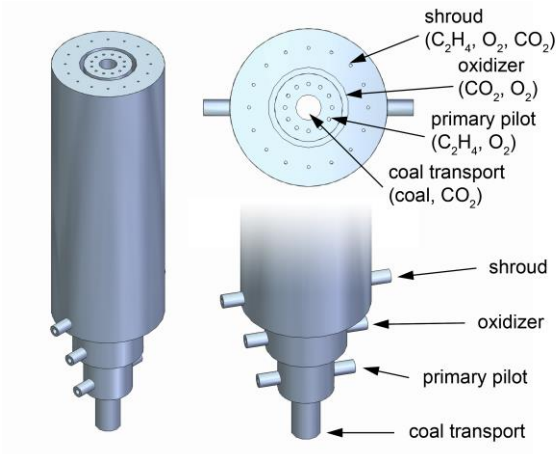

Figure 2: The pulverized-coal burner used for the experiments

Pulverized coal particles leaving through the central hole of the burner were ignited by a primary pilot ethylene flame. The premixed ethylene-oxygen mixture was flowing out through the annular hole pattern surrounding the central hole. The flow velocity of the premixed gas was reasonably high (around 2 m/s at the burner surface), which facilitated early contact with the coal particles. Additional CO_2 and/or O_2 can be optionally added through the annulus surrounding the primary pilot hole pattern, but this stream was not used in the current study. The outermost hole pattern was used for a stabilizing and shrouding ethylene-oxygen flame, which reduced the heat loss of the flame and prevented air intrusion in the studied region of the flame. This shrouding flame also increased overall flame length. Table 1 gives the relevant dimensions of the burner.

Table 1: Relevant dimensions of the burner used for experiments (refer to Figure 2 for explanations)

	# of holes	ID, mm	OD, mm
coal transport	1	7.94	-
primary pilot	12	1.18	-
oxidizer (annulus)	1	21.18	24.89
shroud	16	1.016	-

The burner was enclosed by an acrylic glass box. Flue gas left the box through the top, which was connected to a flue gas disposal system. Air was entrained into the encasing box from below the burner, which was positioned in a circular opening on the bottom side of the encasement. The whole setup was mounted on one end of an optical table, which also served as a basis for mounting the optical diagnostics system.

Operating conditions

The burner described above allowed for realizing a diverse range of combustion conditions, including simulated oxy-firing conditions. The operating conditions tested in this work had been set up so that some of the factors affecting tar formation and behavior – heating rate, the thermal properties of the atmosphere, equivalence ratios, gas and particle velocities – were separated as much as possible.

In essence, five different cases were tested. The five cases represented pure oxy-fuel conditions with 21% O_2 in the mixture, pure air firing conditons, oxy-fuel conditions with an O_2 concentration such that the mixture has similar thermal properties than those of air (this means roughly 34% O_2 in the mixture, in agreement with [5]). Two more cases representing intermediate mixture compositions between pure oxy- and air firing were also tested. The flow rates of gases injected into the shroud flame and the pilot flame were constant – the shroud flame had 2 SLPM CO_2, 0.5 SLPM O_2 and 0.3 SLPM C_2H_4, while the pilot flame was a fuel-rich flame produced by 0.3 SLPM C_2H_4. Table 2 shows the flow rates in the coal transport stream for the different test cases.

Table 2: Flow rates in the coal transport stream for the five different test cases

	O_2	N_2	CO_2
pure oxy-firing	0.318	0	1.2
pure air firing	0.318	1.2	0
high O2 oxy-firing	0.51	0	0.98
intermediate oxy-air I	0.318	0.4	0.8
intermediate oxy-air II	0.318	0.8	0.4

Optical diagnostics

The purpose of the realized optical diagnostics system was to allow for the direct observation of tar trails and other secondary products in the flame, while providing information about the velocities, temperature, radiation profile and CO2 distribution of the studied flame.

A synchronized system of three different imaging methods was constructed – the system included a particle imaging velocimetry (PIV) system, a high-speed mid-wave infrared (MWIR) camera and a high-speed visible camera equipped with a long-distance microscope.

The purpose of the PIV system was to map gas velocities in the flame, particularly in the vicinity of coal particles. From the acquired PIV images, it was also possible to deduce qualitative information about the distribution of coal particles and tar trails. The PIV system was based on a LaVision-made PIV setup that included a LaVision Imager Pro X 4M dual-framing camera and a New Wave Research Solo PIV type dual-head Nd-YAG laser. The frame rate of the system was 10 Hz, while the interframe time was around 300 μs.

The infrared camera utilized for radiation imaging was a FLIR SC6703 type high-speed MWIR camera. The camera was equipped with a narrow-band bandpass filter centered around 4250 nm – this band corresponded to an emission band of CO_2. The integration time of the camera was 100 ns. The camera operated at a frame rate of 10 Hz. The MWIR camera was calibrated by using a controllable temperature black-body cavity – the calibration allowed for the quantitative measurement of radiative flux within the passband.

The purpose of the high-speed visible camera (Photron APS-RX) was to directly observe the formed tar and secondary products in the flame. The observation was based on the principles of shadowgraphy – a pulsed, ultra high-power LED light source was put in-line with the camera, behind the flame, so that the crisp shadows of the observed condensed-phase objects formed in the focal plane of the system. The high-speed visible camera was synchronized with the other devices, such that the effective frame rate was 10 Hz, however the high-speed visible observation was also carried out in a dual-framing fashion, similar to the PIV measurements. The dual-framing mode allowed for the direct measurement of particle and condensed-phase object velocities in the focal plane. The high-speed camera was equipped with a long-distance microscope (Infinity K2/SC), so that a point resolution of around 10 μm was achieved in the field of view.

The synchronization of the three subsystems was carried out by using two programmable digital signal generators and the programmable timing unit that was part of the PIV system. The timing scheme was designed so that the different lighting units (the pulsed laser and LED) did not interfere with each other – in other words, no laser light was recorded in the shadowgraphs and no LED light was recorded in the PIV images. The MWIR imaging coincided with the PIV images, but there was no laser output in the spectral range of the MWIR camera. The time delay between the high-speed shadowgraphs and the images of the other two cameras was on the order of a few μs – this delay was so minimal that there was no conceivable shift between the corresponding PIV, MWIR and high-speed shadow images.

The spatial arrangement of the setup can be seen in Figure 2.

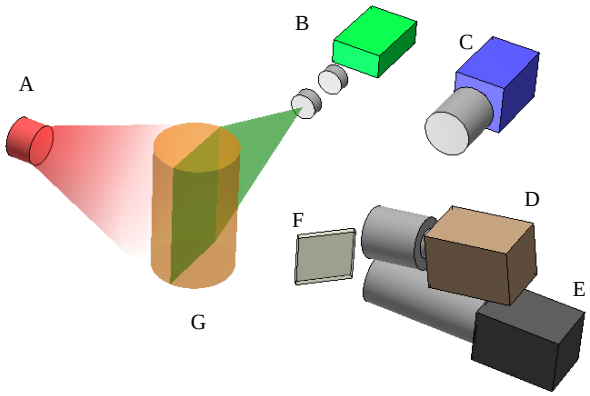

Figure 2: The spatial arrangement of cameras and light sources. A – pulsed LED, B – pulsed Nd-YAG laser, C – PIV camera, D – MWIR camera, E – high-speed camera with long-distance microscope, F – beam splitter, G – flame

Experimental results

Figures 3-5 show the results of the pure air- and oxy-firing cases and the O_2-enriched oxy-firing case.

In the pure air-fired case, devolatilization readily happened at the base of the flame, quickly after the particles entered the pilot flame and began heating up. The volatile bubbles are easily recognizable in the shadowgraphs from 5 mm above the burner. These volatile clouds then formed a condensed phase – opaque trails were observed between 10-20 mm above the burner surface. Further upstream, the formed condensed material burned in a heterogeneous, char oxidation process – the irregularly shaped, elongated and breaking up shapes were indicative of this in the shadowgraphs.

Looking at the pure oxy-firing case, the most striking result is the total suppression of the formation of secondary pyrolysis products and even volatile bubbles. The possible causes of this effect can be the lower flame temperature due to the higher heat capacity of the combustion atmosphere (79% CO_2 + 21% O_2 mixture), the higher gas velocities and the chemistry changes in the flame. The first hypothesis may seem supported by the lower MWIR flux of the flame relative to that of the pure air-fired case, even though there was much more CO_2 in the oxy-fired flamed. PIV results indicated higher velocities along the flame centerline, which may have been caused by the increased viscosity and less spreading of the flame, as compared to the air-fired flame. Chemistry effects may include lowered devolatilization rates due to the higher partial pressure of CO_2 in the atmosphere surrounding particles. Again, no secondary products (volatiles, tar or visible soot particles) were observed in any location of this flame.

When using a CO_2/O_2 mixture with an O_2 concentration so that the thermal properties of the

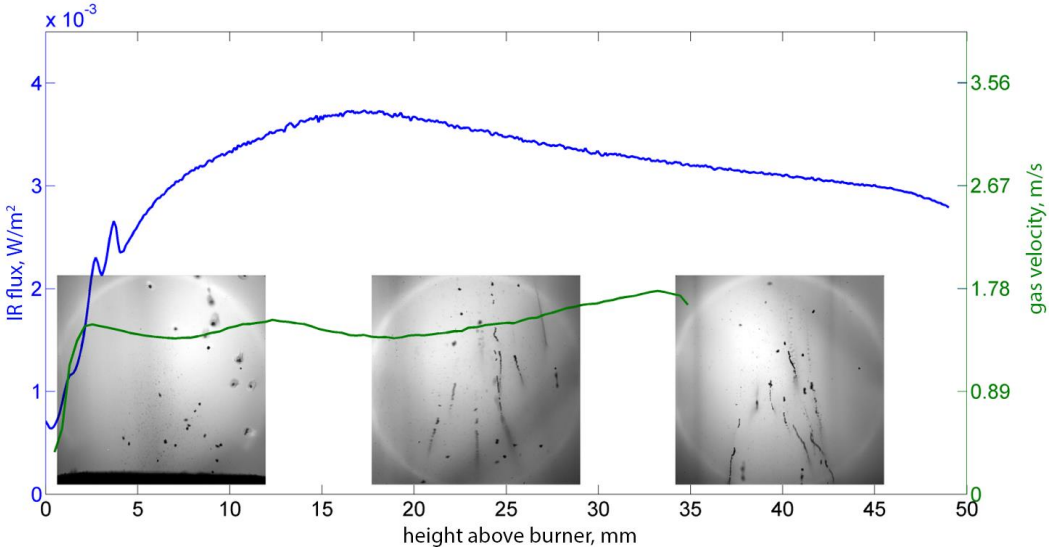

Figure 3: MWIR flux and gas velocity profiles in the centerline of the flame, along with shadowgraphs of coal particles and secondary products. The results showed are from the pure air firing case.

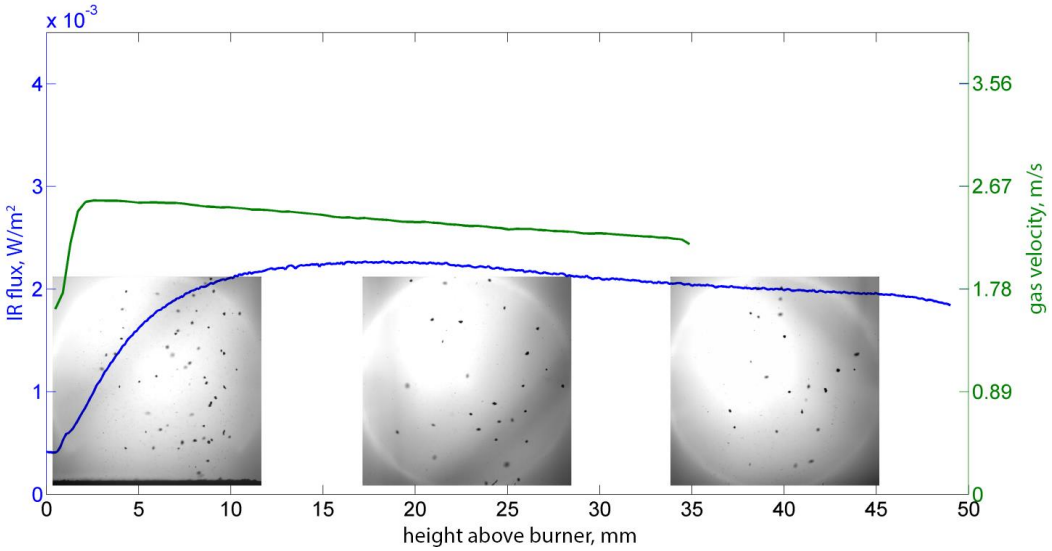

Figure 4: MWIR flux and gas velocity profiles in the centerline of the flame, along with shadowgraphs of coal particles and secondary products. The results showed are from the pure oxy-firing case.

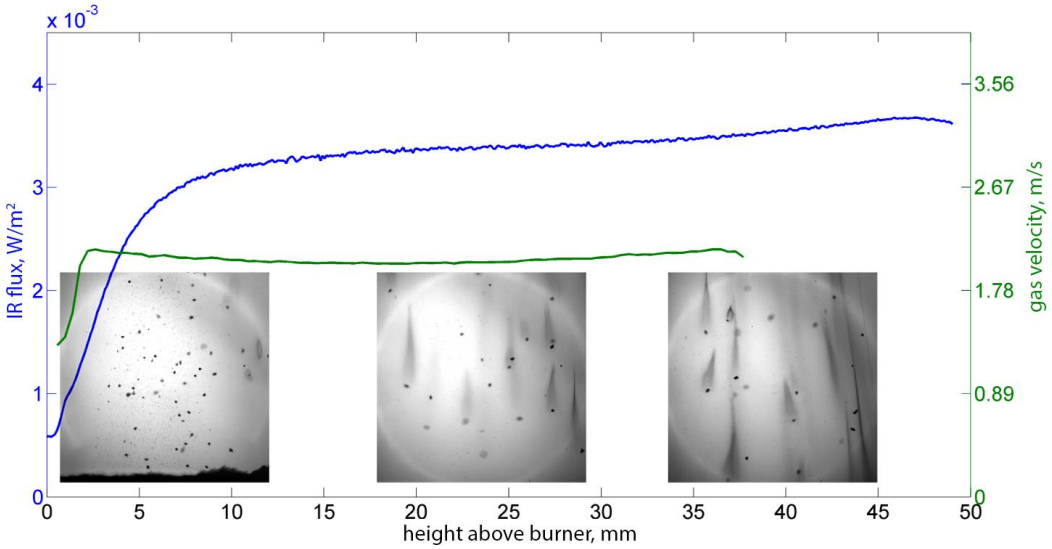

Figure 5: MWIR flux and gas velocity profiles in the centerline of the flame, along with shadowgraphs of coal particles and secondary products. The results showed are from the enriched oxy-firing case.

mixture approximated those of air [5], some volatile bubbles were observed between 20 and 30 mm above the burner in the flame. These bubbles formed typical drop-shaped envelopes further upstream – these shapes are likely the precursors of tar-tails and are created from volatile bubbles being sheared by the gas co-flow. Some very elongated, intact trails formed in the fuel-rich outer regions of this flame. The radiation profile of this case was similar in magnitude to that of the first, air-fired case – the agreement suggests that the thermal properties of the combustion atmosphere were indeed similar to those of air. Gas velocities were between those observed in the purely air- and oxy-fired cases, indicating that the viscosity of the gas mixture was indeed a factor strongly determining final gas velocities. Since the apparent temperature of the flame was similar to that of the air-fired flame, it would seem reasonable to suggest that chemistry changes caused by switching to oxy-firing play a not-so-insignificant role in tar-tail formation. This conclusion however must first be validated by independent (in bands avoiding CO_2 emission lines) temperature measurements in these flames.

The in-between cases of mixed oxy-air flames showed an almost linear transition between pure oxy- and air fired behavior in terms of tar-tail formation.

Conclusion

Imaging-based optical diagnostics were carried out on a laboratory-scale oxy-coal flame to directly observe the formation and behavior of secondary pyrolysis products, namely volatile clouds, tar and soot trails. It was found that the formation of many well-defined observed tar-tails in the air-fired case was suppressed completely when switching to conventional oxy-firing. When using an O_2-enriched oxy condition, some condensed matter began to appear in the flame, although in much reduced quantities relative to air firing. Observed trends in MWIR flame radiation and gas velocities suggest that local chemistry may play a role in the suppression of formation of tar in oxy-coal flames, along with the effect of altered heating rates and lastly, altered gas velocity profiles.

Acknowledgment

This research was carried out in the framework of the Center of Excellence of Sustainable Resource Management at the University of Miskolc. This material is based upon work supported by the Department of Energy under Award Number DE-NT0005015. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

References

[1] T. Wall, Y. Liu, C. Spero, L. Elliott, S. Khare, R. Rathnam, F. Zeenathal, B. Moghtaderi, B. Buhre, C. Sheng, R. Gupta, T. Yamada, K. Makino, J. Yu, Chemical Engineering Research and Design 87(8) (2009) 1003-1016

[2] J. Zhang, K. E. Kelly, E. G. Eddings, J. O. L. Wendt, Proc. Comb. Inst 33(2) (2011) 3375-3382

[3] C. Galletti, E. Giacomazzi, S. Giammartini, G. Coraggio, L. Tognotti, Energy&Fuels 27 (2013) 2732-2740

[4] E. Marek, B. Swiatkowski, Applied Thermal Engineering 66 (2014) 35-42

[5] J. Riaza, R. Khatami, Y. A. Levendis, L. Álvarez, M. V. Gil, C. Pevida, F. Rubiera, J. J. Pis, Comb. Flame 161 (2014) 1096-1108

[6] C. R. Shaddix, S. Molina, Proc. Comb. Inst. 32 (2009) 2091-2098

[7] R-G. Kim, D. Li, C-H. Jeon, Experimental Thermal and Fluid Science 54 (2014) 212-218

[8] S. M. Choi, KSME Journal 4(1) (1990) 71-77