A Passive Mid-Infrared Sensor to Measure Real-Time Particle Emissivity and Gas Temperature in Coal-Fired Boilers and Steelmaking Furnaces

by

Salvador Rego-Barcena

A thesis submitted in conformity with the requirements for the degree of Doctor of Philosophy
Department of Mechanical and Industrial Engineering
University of Toronto

© Copyright by Salvador Rego-Barcena 2008
A Passive Mid-Infrared Sensor to Measure Real-Time Particle Emissivity and Gas Temperature in Coal-Fired Boilers and Steelmaking Furnaces

Salvador Rego-Barcena

Doctor of Philosophy

Department of Mechanical and Industrial Engineering
University of Toronto
2008

Abstract

A novel technique for measuring gas temperature and spectral particle emissivity in high-temperature gas-particle streams is presented. The main application of this optical sensor is to improve the process control of batch unit operations, such as steelmaking furnaces. The spectral emission profile of CO and CO₂ and the continuous particle emission in the 3.5 to 5 µm wavelength region was recorded and analyzed in real time with a low-resolution passive sensor. The sensor consisted of light collecting optics, a dispersion element (grating spectrometer) and a 64-pixel pyroelectric array. Wavelength and radiance calibrations were performed. The temperature of the gas-particle medium \( \left( T_{g+p} \right) \) followed from the least-squares minimization of the difference between the measured radiance in the 4.56-4.7 µm region –which saturates due to the large CO₂ concentrations and path lengths in industrial furnaces– and the corresponding blackbody radiance. Particle emissivity \( \varepsilon_p \) was calculated at 3.95 µm from an asymptotic approximation of the Radiative Transfer Equation that yields the emerging radiance from a semi-infinite particle cloud. The major source of error in the magnitude of \( T_{g+p} \) and \( \varepsilon_p \) could come from particle scattering. Through the method of embedded invariance an expression was
developed to estimate the lowering effect of particle size and volume fraction on the saturation of the 4.56-4.7 \( \mu \text{m} \) \( \text{CO}_2 \) emission region. An iterative procedure for correcting the values of the gas-particle temperature and particle emissivity was applied to the datasets from the two industrial tests. Results from the measurement campaigns with the infrared sensor prototype at two full-scale furnaces are presented. A proof-of-concept test at a coal-fired boiler for electricity production was followed by more extensive measurements at a Basic Oxygen Furnace (BOF) for steelmaking. The second test provided temperature and particle emissivity profiles for eight heats, which highlighted the simplicity of the technique in obtaining in-situ measurements for modeling studies. Through the analysis of the particle emissivity profile in the BOF and the definition of a new variable –the minimum carbon time– a novel end-point strategy to stop the injection of high-purity oxygen during low-carbon heats in BOF converters was proposed.
Acknowledgments

The story that is about to unfold in front of your eyes, dear amici scientiarum, has its source in Canada’s capital. The year was 2004 and the author was completing his Master’s degree at the University of Ottawa. There, Bill Hallett, chair of mechanical engineering, suggested that I contact Murray Thomson as a possible supervisor for my doctorate. It was Bill Hallett himself who had encouraged me on in my academic studies, a decision for which I remain gratefully indebted to him.

If the end of a thesis provides any excuse for one to speak his mind freely, I may say that Murray Thomson turned out to be one of those larger-than-life characters that one is glad to chance upon: a busy professor, father of four –his wife, Esme, also an academic–, optimistic and fun-loving. I feel that I have learned a great deal under his supervision, and not only about sensors, spectroscopy and industrial research. My hope is that future research projects may provide an opportunity to further this professional collaboration and friendship.

I have been very fortunate to count on professors Susan McCahan and Andreas Mandelis as members of my academic committee. I would also like to highlight Susan’s direct help through the Prospective Professors in Training Program, which allowed me to tackle with realism and enthusiasm the non-negligible tasks of applying to faculty positions and learning about teaching in engineering.

The department of Mechanical and Industrial Engineering has been a stimulating environment thanks to its great people. Four professors come readily to mind as outstanding individuals whom I have met here: Charles Ward (and his wife Barbara), Markus Bussmann, Pierre Sullivan and David James. Some were teachers to me, others mentors, all remain good friends.

The success of the infrared sensor project hinged on the unwavering support offered by our industrial partners, Tenova Goodfellow and Unisearch Associates. I collaborated more closely with people at Tenova and would therefore like to thank Howard Goodfellow, Joe Maiolo, Bernie Goldberg and Mohamed Boutazakhti for their interest and help as the infrared sensor technology was developed. I would also like to thank patent lawyer Gary Travis (Riches McKenzie Herbert LLP) for his help with the two patent applications that emerged from this
work. The financial assistance of Sustainable Development Technology Canada and of the Province of Ontario (OGS and OGSST scholarships) is also gratefully acknowledged.

A big eye-opener as I launched on this project was to meet the staff at the Ontario Centres of Excellence (OCE). The dream-team formed by Dan McGillivray, Michael Fagan, Balinder Rai and Leanne Gelsthorpe left a deep impression on my idea of how to form successful research collaborations. Thank you for making me part of your delegation at the GLOBE conference in Vancouver in 2006. I gratefully acknowledge OCE’s Professional Awards that allowed me to attend the Combustion Institute meetings in Heidelberg, Germany, in 2006 and in Banff, Alberta, in 2007. Thanks go to Roxy Hamilton, who led the Value Added Personnel Program at OCE for the fruitful networking and useful business and communication skills that were offered through that program.

If the help of our industrial partners opened the door to the industrial facilities where we could test the infrared sensor, it was the colleagues in the Combustion Group at the University of Toronto who helped in different ways in the building and testing of the two prototypes. It is fair to say that without the help of Rebecca Saari, Reza Mani, Sameh El-Batroukh and Fut Yang this project would not have been as successful as it has been.

I am very grateful to professor Adel Sarofim (University of Utah), whom I met at an American Flame Research Committee conference in Atlanta, Georgia, in 2005, for pointing me to the use of RADCAL. This one-dimensional solver of the Radiative Transfer Equation (RTE) proved invaluable in the analysis of the data from the infrared sensor.

It was a terrific help to count on professor Bruce Hapke (University of Pittsburgh), who kindly reviewed the derivation of a new asymptotic solution of the RTE proposed here. Dr. Champion Chigwedu (Arcelor Mittal’s Burns Harbour facility) offered willing and instrumental help in the understanding of steelmaking furnaces. Thanks go to professor Alex McLean (University of Toronto) for his encouraging comments on the proposed process control strategy for oxygen injection in steelmaking furnaces.

I greatly enjoyed the friendship and support that I found among fellow graduate students at the University of Toronto. I would like to mention by name those closest to me for being part of the
Combustion Group: Mani Sarathy (and his wife Nimmi), Tom Tzanetakis, Amir Aliabadi and Qingan Zhang.

The staff at the Mechanical and Industrial Engineering department has been very reliable and helpful. Thanks go to Brenda Fung and Lorna Wong, Joe Baptista, Sheila Baker, Teresa Lai-Ho, Oscar del Rio, Joey Wong, Tom Bernreiter, Len Roosman, David Esdaile, Tai Tran Do and Wanda Grabiec for making the little things easy.

These four years in Toronto have been very fruitful in many other ways, among other things for the quality of the people whom I met at Ernescliff College. All of them deserve my special thanks for their inspiring example and friendship.

The most important acknowledgement belongs to my parents, María del Carmen and Juan Ramón, to my sister María and my brothers Juan, José María, Carlos (and his fiancée Carmen), Jesús (in pace) and Pablo. Thank you! This thesis is dedicated to my parents.
# Table of Contents

Acknowledgments.......................................................................................................................... iv  
Table of Contents.......................................................................................................................... vii  
List of Tables ................................................................................................................................ x  
List of Figures ................................................................................................................................ xi  
List of Appendices ....................................................................................................................... xiv  
Nomenclature................................................................................................................................ xv  
Acronyms..................................................................................................................................... xix  
1 Introduction............................................................................................................................... 1  
2 The Infrared Sensor: Description and Proof-of-Concept Test at a Coal-Fired Boiler ............. 8  
   2.1 Introduction.......................................................................................................................... 8  
   2.2 Methodology ...................................................................................................................... 12  
      2.2.1 Instrument Description .............................................................................................. 12  
      2.2.2 Wavelength and Radiance Calibration ...................................................................... 13  
      2.2.3 Description of IR Spectra ......................................................................................... 15  
      2.2.4 Method of Temperature Measurement .................................................................... 19  
      2.2.5 Method of Particle Emissivity Measurement ............................................................ 20  
   2.3 Results ............................................................................................................................. 24  
      2.3.1 Industrial Test at a Coal-Fired Boiler (Nanticoke) .................................................... 24  
      2.3.2 Raw Data Analysis .................................................................................................... 25  
      2.3.3 Temperature and Emissivity Results ....................................................................... 26  
   2.4 Discussion ......................................................................................................................... 28  
      2.4.1 Error and Uncertainty Analysis ................................................................................. 31  
   2.5 Conclusions ....................................................................................................................... 34  
   2.6 References......................................................................................................................... 35
3  An Analytical Expression and Iterative Procedure to Check for Particle Scattering Effects on the Gas Temperature and Particle Emissivity Retrieved by the IR Sensor .............................. 38
   3.1  Introduction .................................................................................................................. 38
   3.2  Effective Emissivity of a Semi-Infinite Medium with Gas and Micron-Sized Particles.. 41
       3.2.1  Derivation .......................................................................................................... 42
   3.3  Discussion .................................................................................................................... 48
       3.3.1  Application to Temperature and Emissivity Data from a Coal-Fired Boiler ...... 51
       3.3.2  Accounting for Particle Effects with an Estimate of $\chi$ ................................. 52
   3.4  Conclusions ............................................................................................................... 53
   3.5  References .................................................................................................................. 54

4  Real Time Gas Temperature and Particle Emissivity at a Basic Oxygen Furnace for Steelmaking .................................................................................................................. 57
   4.1  Introduction ................................................................................................................ 57
   4.2  Methodology .............................................................................................................. 59
       4.2.1  Temperature and Particle Emissivity Retrieval ............................................... 59
       4.2.2  Calculating $\chi$ to Validate the Assumption of Saturation in the Gas-Particle Region ................................................................................................................... 62
   4.3  Experimental ............................................................................................................. 65
   4.4  Results ......................................................................................................................... 68
   4.5  Discussion .................................................................................................................. 71
       4.5.1  Analysis of the Off-Gas Temperature Data ......................................................... 71
       4.5.2  Analysis of Particle Emissivity Data ................................................................. 74
       4.5.3  Assumptions in the Retrieval Algorithms ......................................................... 78
       4.5.4  Particle Scattering Effects and Uncertainty Analysis ....................................... 79
   4.6  Conclusions ............................................................................................................... 82
   4.7  References .................................................................................................................. 83

5  Process Control Implications of Measuring Real-Time Gas Temperature and Particle Emissivity in a Basic Oxygen Furnace .................................................................................. 86
List of Tables

Table 4-1. Absorption coefficient $\kappa_g$ [m$^{-1}$] at 4.60 µm for a 2.87 m gas column at several compositions (vol-%) and temperatures from the EM2C model – p. 63

Table 4-2. Bath properties from the seven heats of the field trials – p. 67

Table 4-3. Data for the steady-state decarburization zone of all heats – p. 71

Table 4-4. Comparison of $\varepsilon_{\text{eff}}$ at 3.95 µm and similar temperatures for relevant oxides and present results – p. 75

Table 4-5. Additives, estimated mass of steel at tap per O$_2$ consumption for heats 1 to 5 – p. 76

Table 4-6. Variables used in the calculation of $\chi$ at $r_p = 2.5$ µm and 5 µm to check for particle scattering effects on $T_{g+p}$ and $\varepsilon_p$ – p. 80

Table 5-1. Aim, first turn down (TD1), hot metal (HM) and initial (In) process data for the eight heats. Compositions are expressed in wt-% – p. 91

Table 5-2. Time at which $\varepsilon_p$ peaked expressed as a percentage of the total oxygen injection time – p. 95

Table A2-1 Bill of materials and suppliers for the building and calibration of the current version of the IR sensor prototype – p. 112
List of Figures

Figure 1-1. Saturated and unsaturated behaviour of the 4.3 $\mu$m CO$_2$ band modeled in RADCAL for different path lengths of a gas column containing 14% CO$_2$ in air at 1 atm and 1500K. Also plotted is the blackbody profile at 1500K (labeled “BB”) – p. 2

Figure 2-1. Main components of the IR sensor – p. 12

Figure 2-2. Sample raw data from the Nanticoke test showing the three spectral regions (a), and results for one scan after radiance calibration was performed (b) – p. 16

Figure 2-3. One-dimensional schematic of the emitted radiation in a boiler – p. 17

Figure 2-4. RADCAL model of saturated radiation emitted by 14% CO$_2$ in air (1 atm) over a path length of 14 m along with three blackbody curves (labeled “BB”) – p. 19

Figure 2-5. The retrieved temperature is the one that minimizes the regression variable $R^2$ – p. 20

Figure 2-6. Simulation of Nanticoke conditions in RADCAL using average values from the proof-of-concept test – p. 23

Figure 2-7. The IR sensor during the industrial test at one of Nanticoke’s coal-fired boilers – p. 25

Figure 2-8. Raw number of counts (a measure of radiance) for two wavelengths (4.60 $\mu$m from the CO$_2$ emission band and 4.30 $\mu$m from the room CO$_2$ absorption band) versus time – p. 26

Figure 2-9. Retrieved physical temperature inside the boiler ($T_{g+p}$) and brightness temperature of the particles ($T_p$) – p. 27

Figure 2-10. Time series of the particle and effective emissivities for the Nanticoke test with the physical temperature plotted on the secondary axis (a). The same emissivity results are presented versus temperature on (b) – p. 28

Figure 3-1. The remote operation of a passive spectroscopic sensor for high-temperature off-gas measurements – p. 38
Figure 3-2. Modeled effective emissivities of an optically-thick particle cloud as a function of the average particle emissivity – p. 39

Figure 3-3. Five changes to the emerging radiance from a semi-infinite gas-particle medium introduced by the addition of a thin layer – p. 43

Figure 3-4. Effective emissivity of a semi-infinite medium of gas and particles for some values of $\chi$ and $\varepsilon_p$ – p. 49

Figure 3-5. Temperature (a) and particle emissivity at 3.95 $\mu$m (b) for different values of $\chi$ after including particle effects – p. 53

Figure 4-1. Simulation of the emitted radiance profile using RADCAL for typical values of the off-gas in the middle of a typical heat (BB = blackbody profile) – p. 61

Figure 4-2. Main components of the IR sensor – p. 66

Figure 4-3. Schematic of the location of the IR sensor on the BOF skirt (a); and a view of the NEMA box with the IR sensor inside (b). The white box is the grating spectrometer and the metal flange on the side attaches the pyroelectric detector to the outlet of the spectrometer (Figure 4-2) – p. 67

Figure 4-4. Sample calibrated profiles from the output of the IR sensor before and after the oxygen lance was stopped (final measuring campaign, heat 7) – p. 68

Figure 4-5. Off-gas temperature ($T_{g+p}$) and particle emissivity ($\varepsilon_p$) retrieved by the IR sensor for the seven heats during the oxygen-blowing period. The data is reported as a moving average every 4 measurement intervals (8.1 seconds) – p. 69

Figure 4-6. Comparison between off-gas temperature data at the mouth of the same converter from a CO$_2$ thermometer (Land Instruments CD1) and the infrared (IR) sensor (heat 2). Data were taken weeks apart – p. 73

Figure 4-7. Linear fits of particle emissivity versus temperature for heats 1 to 5 – p. 76
Figure 4-8. Simulated radiance profiles for different levels of CO$_2$ in the off-gas using RADCAL, and the retrieved temperature from the gas-particle region (4.56-4.7 $\mu$m) – p. 79

Figure 5-1. Main components of the IR sensor (a); Schematic of the location of the IR sensor on the BOF skirt (b) – p. 89

Figure 5-2. Off-gas temperature ($T_{g+p}$; dotted line), particle brightness temperature ($T_p$; solid gray line), particle emissivity ($\varepsilon_p$; solid black line), end of oxygen blow line (solid vertical line) and minimum carbon time ($\tau_{MC} \equiv$ peak emissivity time - end of oxygen time) for eight heats at a 168 tonne BOF converter. Data is time-averaged every 6 cycles or 12.2 s – p. 92

Figure 5-3. Correlation of minimum carbon time ($\tau_{MC}$) and the first turn bath properties –carbon (a), temperature (b), and phosphorus (c)– for four low carbon and four high carbon heats. TD1 = first turn down value. Also shown are the aim values for the four low carbon heats – p. 94

Figure A1-1. RADCAL emission profiles of 10% and 50% CO in air (1 atm, 1600K) with and without (“no BB” lines) broadband particle emission ($\varepsilon_p = 0.4$) modeled by a background temperature of $T_p = 1535K$ – p. 110

Figure A1-2. RADCAL emission profiles at 1 atm and 1600K of 20% CO$_2$ in air, and to mixtures of CO and CO$_2$ (balance = air) with broadband particle emission ($\varepsilon_p = 0.4$) modeled by a background temperature of $T_p = 1535K$ – p. 111
List of Appendices

Appendix 1 The Difficulty of Measuring CO and CO₂ When Particles are Present in Industrial, High-Temperature Gas-Particle Streams with Low-Resolution Emission Spectroscopy in the Mid-Infrared (4-5 µm) – p. 109

Appendix 2 Materials and Operation of IR Sensor and Suggestions for the Hardening of the Prototype – p. 112

Appendix 3 Nature of the Collaboration with other Members of the Combustion Group – p. 115
Nomenclature

\( A_p \)  Particle cross-sectional area [m^2]

\( A_{\text{im}} \)  Target steel temperature for one heat [°C]

\( I_b \)  Planck’s radiance distribution for a blackbody [W/m^2/μm/sr]

\( C \)  Carbon weight fraction in the steel [\text{.}]

\( C_1, C_2 \)  First and second radiation constants

\( e \)  Final emerging angle [rad]

\( e' \)  Initial emerging angle [rad]

\( f_v \)  Particle volume fraction [\text{.}]

\( H \)  Component of the bidirectional reflectance function (spectral) [\text{.}]

\( H_0 \)  First moment of the \( H \) function (spectral) [\text{.}]

\( I \)  Radiance [W/m^2/μm/sr]

\( i \)  Incoming angle [rad]

\( \text{In} \ P \)  Initial phosphorous weight fraction [\text{.}]

\( m_p \)  Particle mass in one heat [kg]

\( N_p \)  Particle number density (in one heat) [m^3]

\( n_p \)  Number of particles (in one heat) [\text{.}]

\( P \)  Phosphorus weight fraction in the steel [\text{.}]

\( P\% \text{ reduct.} \equiv (\text{In} \ P - \text{TD1} \ P)/\text{In} \ P \) [\text{.}]

\( R^2 \)  Regression variable [(W/m^2/μm/sr)^2]
\( r \)  Bidirectional reflectance function (spectral) [\( . \)]

\( r_p \)  Particle radius [\( \mu m \)]

\( S \)  Path length across the gas-particle column [m]

\( T \)  Temperature [K]

\( T_{\text{Diff}} \equiv TD1 \ T - \text{Aim} \ T [K] \)

\( TD1 \ C \)  Carbon weight fraction in the steel at first turn-down [\( . \)]

\( TD1 \ P \)  Phosphorus weight fraction in the steel at first turn-down [\( . \)]

\( TD1 \ T \)  Steel temperature at first turn-down [K]

\( T_{g+p} \)  Off-gas temperature [K]

\( T_p \)  Brightness temperature from the particle wavelength region [K]

\( V_{\text{offgas}} \)  Total off-gas volume produced in one heat [m\(^3\)]

\( V_{O2} \)  Total lance oxygen volume consumed in one heat [m\(^3\)]

\( V_p \)  Total volume of particles in one heat [m\(^3\)]

\( X \equiv 2\pi \ r_p / \lambda \)  Particle size parameter [\( . \)]

Greek symbols

\( \beta \)  Extinction coefficient [m\(^{-1}\)]

\( \chi \equiv \kappa_g / N_p A_p \)  Parameter used to estimate particle effects [\( . \)]

\( \Delta I \)  Change in the outgoing radiance (spectral) [W/m\(^2\)/sr/\( \mu m \)]

\( \Delta \tau \)  Optical thickness of thin layer (spectral) [\( . \)]
$\Delta \tau_{g}, \Delta \tau_{p}$  Gas and particle components of thin layer’s optical thickness (spectral) [\(\cdot\)]

$\Delta z$  Thickness of thin layer [m]

$\varepsilon_{\text{eff}}$  Effective emissivity of a single- or double-component medium (spectral) [\(\cdot\)]

$\varepsilon_{p}$  Particle emissivity at 3.95 µm (spectral) [\(\cdot\)]

$\varepsilon_{\text{tot}}$  Total particle emissivity [\(\cdot\)]

$\gamma$  Albedo factor of the particle medium [\(\cdot\)]

$\kappa_{g}$  Gas absorption coefficient (spectral) [m\(^{-1}\)]

$\kappa_{p}$  Particle absorption coefficient (spectral) [m\(^{-1}\)]

$\lambda$  Wavelength [µm], wavelength-dependent variable

$\mu$  Cosine of the emergence angle ($e$) of the emitted radiation [\(\cdot\)]

$\mu', \mu_{0}$  Cosine of $e$, $e'$ and $i$ [\(\cdot\)]

$\Omega_{e}, \Omega_{e'}, \Omega_{i}$  Infinitesimal solid angles associated with $e$, $e'$ and $i$ [sr]

$\tau$  Optical thickness (spectral) [\(\cdot\)]

$\tau_{\text{MC}}$  Minimum carbon time [s]

$\omega_{p}$  Particle single-scattering albedo (spectral) [\(\cdot\)]

$\omega_{p+g}$  Medium single-scattering albedo (spectral) [\(\cdot\)]

$\sigma_{g}$  Gas scattering coefficient (spectral) [m\(^{-1}\)]

$\sigma_{p}$  Particle scattering coefficient (spectral) [m\(^{-1}\)]
Subscripts

\[ g \quad \text{gas-phase property} \]

\[ g+p \quad \text{off-gas property (containing gases and particles) / related to the gas-particle wavelength region (4.56-4.70 \, \mu m)} \]

\[ p \quad \text{particle-only property / related to the particle-only wavelength region (3.8-4.1 \, \mu m)} \]

source temporary IR Emitter used in radiance calibration for proof-of-concept test

1…5 Index for changes in the outgoing radiance
Acronyms

Aim     Target value for a property to be achieved at the end of a heat
BOF    Basic Oxygen Furnace for steelmaking
CFD    Computational Fluid Dynamics
EAF    Electric Arc Furnace for steelmaking
HM     Hot metal property
RTE    Radiative Transfer Equation
TD1    First turn-down property
1 Introduction

This thesis documents the retrieval theory and experimental results with a remote, optical sensor for industrial furnaces. The structure follows the four publications that capture the main contributions that the author hopes to make to the field of combustion diagnostics.

This introduction will provide the overall context for the so called “infrared or IR sensor” and highlight the common thread that unites all chapters. By context is understood the definition of the conceptual building blocks and objectives or deliverables that give meaning to this work. The thesis ends with a conclusions chapter where the main contributions, their novelty and ideas for future work are outlined.

This work rests on two well-known concepts in radiation heat transfer. First, at high temperatures all solids emit radiation continuously over all wavelengths, while some gases emit only over certain wavelength bands. Secondly, the maximum thermal radiation that may be emitted is capped by Planck’s distribution for a blackbody (Equation 2-4), which is attained by solids with an emissivity of unity or by gases when their emission is said to be saturated. Since a solid’s emissivity is spectral (that is, it varies with wavelength) and gases are intrinsically spectral emitters, this blackbody behaviour may be only observed over certain regions of the spectrum. A good example is white paint, a gray body with very low emissivity ( < 0.2) in the visible region (0.4 to 0.7 µm), but that behaves more like a blackbody in the mid-IR (λ > 3 µm) with emissivities around 0.9 [1]. Gases become saturated over some of their emission bands if either their concentration or the size of the gas column is large enough. Figure 1-1 shows the progressive saturation of the 4.3 µm CO₂ band as the path length increases. Already at a path length of 0.5 m the core of the band matches the blackbody profile at the same temperature
(labeled “BB”). The conditions for saturation are found in many full-scale furnaces, such as the coal-fired boiler of Chapter 2 or the basic oxygen furnace (BOF) of Chapters 3 and 4. Figure 1-1 was generated with RADCAL [2], a one-dimensional solver of the Radiative Transfer Equation (RTE) that has been very useful for this doctoral project.

![Graph showing saturated and unsaturated behavior of CO2 band](image)

Figure 1-1. Saturated and unsaturated behavior of the 4.3 \( \mu \text{m} \) CO\(_2\) band modeled in RADCAL [2] for different path lengths of a gas column containing 14% CO\(_2\) in air at 1 atm and 1500K. Also plotted is the blackbody profile at 1500K (labeled “BB”).

The regions surrounding the core of a gas band are called wings and they typically do not saturate. A consequence of this is that a saturated band will not look like a perfect step function on both sides of its core. The gradual slope in the wings of gaseous bands can be observed in the 4.3 \( \mu \text{m} \) CO\(_2\) and 4.5 \( \mu \text{m} \) CO bands of this study.

The region of interest for our work is the mid-IR between 3.5 and 5 \( \mu \text{m} \). For a system consisting of high temperature exhaust gas that entrains solid particles, this spectral region contains gaseous
emission from CO and CO$_2$ superimposed on the particle emission, and another region where there is particle-only emission. The nature of the superimposition between gas and particle emission and the fact that particles scatter IR radiation led to some interesting results (Chapter 3).

We began this project with the motivation of measuring either the absolute concentration of CO and CO$_2$, or at least changes in the ratio of these species in harsh industrial environments, such as power generating stations and steelmaking furnaces. The ultimate objective remains to provide process control practitioners with real-time information about the furnace condition (concentration of CO and CO$_2$ being an example of this type of information) to optimize material and energy inputs to the process. The Combustion Group at the University of Toronto and its industrial partners have had experience with other alternative technologies such as extractive sampling and laser sensors. The IR sensor was a sensible step in overcoming the limitations of the previous approaches: extractive sampling requires maintenance (e.g., the filter that cleans up the gas sample) and does not measure temperature; while laser sensors are still not reliable enough because of beam-attenuation from sudden changes in gas temperature (especially in steelmaking furnaces where they would be most useful) and from particle scattering.

There is one element that was plentifully abundant in these high temperature industrial facilities: visible light and thermal radiation. Thus, the main hypothesis of the IR sensor was to capture and to analyze the emitted radiation from high-temperature gas-particle streams. There is a clear trade-off in this approach: contrary to extractive sampling and laser sensors, data acquisition became relatively straightforward (one only needed to collect the IR radiation with a camera-like device). However, contrary as well to the other two technologies, data analysis and interpretation were not well understood. Questions such as whether CO and CO$_2$ concentrations would be retrievable from IR signature recorded by the sensor were still unanswered.
The riddle has been solved in part. We know now that with the tools employed (low resolution mid-IR emission spectroscopy) one cannot measure CO and CO$_2$ in high-temperature gas streams that entrain particles because of the (typically) gray, continuous background from particles and because the radiation emitted by both species overlap in the 4.6 $\mu$m region (Appendix 1). A less problematic question (not explored here) would be to quantify the concentration of these gases if they were present alone. A more researched topic would be to retrieve their concentration in a high temperature gas without entrained particles.

In figuring out the CO-CO$_2$-particle dilemma some original answers were found: while it is not possible to retrieve CO and CO$_2$ concentrations, the gas temperature ($T_{g+p}$) and the particle emissivity ($\varepsilon_p$) in the mid-IR can be retrieved in real-time from the radiance versus wavelength profile recorded by a relatively simple experimental set-up. And further analysis of this data showed its usefulness for improved process control in batch unit operations.

Thankfully this project has counted with generous industrial and institutional support. This collaboration made it possible to test the IR sensor at Nanticoke and U.S. Steel Canada (formerly Stelco). Nanticoke Generating Station, on the north shore of Lake Erie, is the largest coal-based electrical power producer in North America. One of its eight boilers was the site of the first industrial test with the IR sensor (Chapter 2). One of the three BOF vessels at U.S. Steel Canada in Hamilton provided a longer dataset from which real-time temperature and particle emissivity profiles for eight heats (a batch process to refine pig iron into steel) were measured (Chapter 4) and analyzed for their potential to provide a reliable control strategy for oxygen injection (Chapter 5). Our involvement with industrial partners guided the building of a prototype that could perform the data analysis calculations in real-time, and thus avoid post-processing of the results. Therefore an analytical solution to estimate the particle emissivity was developed, but the correction for particle scattering effects remains a post-processing task as explained in Chapter 3.
The objectives for this thesis are:

1. To develop a methodology to measure gas temperature and particle emissivity in real-time (every two seconds) in high-temperature industrial exhaust streams.

2. To build and calibrate the hardware needed to measure radiance versus wavelength in the 3-5 \( \mu \text{m} \) region using as a building block a 64-pixel linear array pyroelectric detector. A comprehensive list of materials and suggestions for the hardening of this prototype into a commercial prototype appear in Appendix 2.

3. To describe the algorithms to retrieve gas-particle temperature \((T_{g+p})\) and particle emissivity \((\varepsilon_p)\).

4. To provide the theoretical basis to correct the magnitude of \(T_{g+p}\) and \(\varepsilon_p\) due to particle scattering effects and to establish the conditions that warrant this correction.

5. To propose a process control strategy for high-purity oxygen injection in a BOF facility based on the analysis of the data from the IR sensor.

The second chapter summarizes the components of the first IR sensor prototype and describes the proof-of-concept test at Nanticoke. Some of the main ideas are the wavelength and radiance calibration; the nature of the three spectral regions recorded by the sensor (particle-only emission, room CO\(_2\) absorption, high temperature gas-particle emission); the use of the saturated gas-particle emission region to retrieve the gas-particle temperature by least-squares minimization by comparing blackbody radiance with the measured radiance; the derivation of analytical formula to calculate the particle emissivity in the mid-IR based on the method of embedded invariance (this was very useful for the later integration of the data acquisition and retrieval algorithm components into one software package); the distinction between the particle brightness temperature \((T_p)\) and physical temperature of the gas-particle stream \((T_{g+p})\), and between the effective emissivity of the gas-particle stream \((\varepsilon_{eff})\) and the particle emissivity \((\varepsilon_p)\).
The third chapter summarizes the theoretical work behind the estimation of particle scattering effects, which were already announced in Chapter 2. These scattering effects are a function of two particle properties—their number density ($N_p$) and average cross-sectional area ($A_p$)—and the gas absorption coefficient ($\kappa_g$), which were combined in a dimensionless new parameter ($\chi \equiv \kappa_g/N_pA_p$) as a convenient tool to estimate whether to correct for particle scattering effects. The main features are the development of an original analytical expression for the emerging radiance from a semi-infinite column of gas and particles; the establishment of threshold values of $\chi$ and initial $\varepsilon_p$ that render the temperature retrieved from the gas-particle region lower than the actual physical temperature, and the particle emissivity higher than it should; an iterative procedure to correct (a posteriori) temperature and emissivity values; the successful application of this procedure to the data from Chapter 2 since it was discovered that particle scattering effects did have an impact on the magnitude of the retrieved temperature and emissivity from the Nanticoke test.

The fourth chapter relies on the well-established methodology outlined in Chapters 2 and 3 to face a new diagnostic challenge: the exhaust gas from a BOF facility where pig iron is oxidized to produce steel. The advantage of this system relative to the coal-fired boiler is its batch nature, which allowed us to observe clear variations in the retrieved variables since the boiler at Nanticoke had been a fairly steady-state system. The gas temperature and particle emissivity from eight heats (or batches of pig iron) were reported. In themselves $T_{g-p}$ and $\varepsilon_p$ are useful variables. Gas-particle temperature can describe boundary conditions in computationally fluid dynamics (CFD) codes and particle emissivity is a required parameter in the Radiative Transfer Equation.

Chapter 5 explores the transient nature of the temperature and emissivity traces as the basis of a plausible control strategy for oxygen injection into the BOF (high-purity oxygen is the main
agent in the decarburization of pig iron to produce steel). Large benefits would ensue from such as a strategy (increased productivity and reduced environmental impact being the two most significant). An original contribution of this chapter is the definition of a new variable (minimum carbon time, $\tau_{MC}$) which measures the time between a consistent peak in $\varepsilon_p$ towards the end of the heat and the time at which the oxygen injection ended. The correlations of $\tau_{MC}$ with the final steel parameters (temperature, carbon and phosphorus content) of the steel bath, referred to as “first turn-down” parameters, provided the quantitative foundation for the proposed end-point control strategy for oxygen injection.

All chapters share a standard nomenclature and formatting style in the naming and cross-referencing of equations, tables and figures, as well as in the bibliography. The Conclusions chapter summarizes the main contributions from this work and suggests future lines of research. As the acknowledgement section makes evident, the author is indebted to his supervisor and colleagues for their help. Appendix 3 describes in detail the nature of this collaboration.
2 The Infrared Sensor: Description and Proof-of-Concept Test at a Coal-Fired Boiler

2.1 Introduction

Instrumentation for combustion diagnostics in full-scale boilers has unique challenges compared to its laboratory counterpart. However industrial diagnostics can provide useful information for process control and modeling studies. This chapter presents a novel technique to measure particle emissivity ($\varepsilon_{p,\lambda}$) from spectral information in the mid-IR.

Laboratory techniques for measuring particle emissivity at high temperatures (e.g., greater than 800K) have been published for coal and char particles [1], fly-ash, slag and fouling from coal samples [2] and the main mineral phases in fly-ash [3]. Their main objective is the determination of emissivity as a thermal property required for heat transfer calculations. Emissivity is calculated as a spectral and directional quantity that is measured according to

$$\varepsilon_{p,\lambda}(T) = \frac{I_{s,\lambda}(T)}{I_{b,\lambda}(T)}, \quad (2-1)$$

where $I_{s}(T)$ is the incident radiation from a layer of pulverized particles and $I_{b,\lambda}(T)$ is the radiation from a blackbody source.

In the abovementioned studies, particle radiation is measured spectrally between 1 and 16.7 $\mu$m with an FTIR or a spectral radiometer. Minimizing the temperature difference between the sample and blackbody source and long measuring times (e.g., in the order of 5+ minutes at each

\footnote{This chapter is based on [20] which appeared in print in 2007. Some improvements have been made to Sections 2.2.1 and 2.4 over [20].}
wavelength interval) are common challenges of laboratory techniques [3]. Moving optical parts and equipment costs preclude the straightforward application of these techniques to in-situ, industrial environments.

Hence in-situ measurements in full-scale, coal-fired boilers are less common [4,5]. In [4] particle emissivities are presented as total emissivities according to spectral measurements between 0.9 and 5 µm and the following equation:

\[
ε_{\text{tot}}(T) = \frac{\int_{λ_1}^{λ_2} ε_{p,λ}(T) I_{b,λ}(T) dλ}{\int_{λ_1}^{λ_2} I_{b,λ}(T) dλ}.
\]  

(2-2)

In [5] emissivity is calculated according to Equation 2-1, from radiometric data from the visible range of the spectrum. Both [4] and [5] present the spatial (vertical) distribution of emissivity within the boiler and comment on the effect that particle size, boiler loading and type of coal have on this property.

In this chapter we present a portable device to measure in-situ and in real time (once every 2 s) particle emissivity based on the emitted radiance of a gas-particle medium between 3.7 and 5 µm. In situations such as the current proof-of-concept test at a coal-fired boiler, the particles are a mixture of fly-ash and unburned coal. Two types of spectral emissivity are calculated. In the first place, by analogy with Equation 2-1, we define an effective emissivity based on the ratio of the emitted radiance from an optically thick mixture of gas and particles to blackbody radiance at the same temperature. Secondly, we derive the particle emissivity as the physical property used in heat transfer calculations. The effective emissivity is larger than the particle emissivity since the emerging radiance for a medium of suspended particles contains the emittance from the
pulverized particles, $\varepsilon_{\lambda}(T)B(\lambda)$, augmented by scattering of radiation in the direction of the instrument caused by the particles themselves.

Particle emissivity is a useful property for CFD studies that include radiative heat transfer from combustion gases and particles in order to calculate the radiative source term in the energy conservation equation for fluid flow. CFD studies of large combustion systems where particles are present require particle properties such as volume fraction, size distribution and emissivity in order to predict the net flux density to the wall. Normally a spectrally averaged solution to the radiative transfer equation is sought, e.g., [6]. This solution typically includes the diffusion approximation (i.e., optically-thick media with constant absorption coefficients for gas and soot) and the total particle emissivity, which appears in the definition of the particle absorption and scattering coefficients (Equations 2-12 and 2-13). A common approximation in modeling coal-fired burners is to make the particle emissivity vary between the emissivity of unburned carbon (1.0) and fly-ash (0.6). For instance, Chui et al. [6] made particle emissivity depend on the fraction of unburned char mass. Other modeling researchers, e.g., [7], further simplified this approach by using an arithmetic average, $\varepsilon_{\text{tot}} = 0.8$, in their study of non-isothermal, non-gray gas and gray particle radiation. Recently Williams et al. [8] used an emissivity of 0.9 for char in their detailed model of coal combustion. One can find in the literature particle emissivity values that would either support the correlation $\varepsilon_{\text{tot}} \in [0.6-1]$ (e.g., [2]) or that suggest lower values (e.g., [4] and the results from the current proof-of-concept test). The main lesson may be that no general formula for particle emissivity will suit all coal-fired systems. Our motivation was to develop a simple technique that is capable of measuring in-situ particle emissivities for modeling studies of specific boilers. Blokh [4] showed how total emissivities within a boiler varied linearly with height in the region above the rows of burners (and in the case low-rank coals, the linearity also extended in to the combustion region). Thus measurements with the current IR sensor at two
or three vertical locations may be enough to characterize the particle emissivity throughout the whole boiler.

Two further advantages are realized by the current method. The retrieved particle emissivity accounts in a single value for the distribution in chemical composition (e.g., char, dielectrics) and size of the particles in the field of view. Although our particle emissivity is spectral ($\lambda = 3.95 \ \mu m$) and heat transfer calculations normally require total emissivity, the data in [3] shows that total emissivity for coal ash and sediment samples between 1 and 15 $\mu m$ at high temperatures (> 1000K) resembles the spectral value near 4 $\mu m$. This wavelength interval covers 98% of the blackbody curve at 1500K. Their data is also useful in that it highlights the low effect of temperature on total emissivity.

Essential to the measurement of particle emissivity with the current technique is the determination of the gas-particle medium temperature. This is accomplished by taking advantage of the blackbody or saturated emission of the 4.3 $\mu m$ band of CO$_2$ in the combustion gas.

Thus, the purpose of this chapter is to present the measurement technique including: (a) the description of the instrument, (b) the retrieval methods for temperature and emissivity, (c) a comparison of our measurements to literature values, and (d) an error and uncertainty analysis. Experimental data was obtained from a proof-of-concept test in a full-scale, coal-fired boiler in Ontario, Canada. The IR sensor was located 10 meters above the top row of burners. A comprehensive survey of particle emissivity throughout the boiler was not the objective of this test.
2.2 Methodology

2.2.1 Instrument Description

The spectral region between 3.7 to 5.0 µm includes continuous radiation from solid particles and discrete radiation from CO₂ in the combustion gas. Thus the IR sensor prototype has been designed to measure radiance versus wavelength in this spectral range.

As is shown in Figure 2-1 there are three main components: a lens tube to collect the radiation, a grating spectrometer to disperse the radiation into different wavelengths, and a linear-array pyroelectric detector.

![Figure 2-1. Main components of the IR sensor.](image)

The lens tube contains a 2.5 cm CaF₂ plano-convex lens (focal distance = 75 mm at 588 nm), a long pass filter (3.60 to 6.89 µm) and a 10 Hz mechanical chopper. The long pass filter is used to block wavelengths between 1.85 and 2.5 µm, whose second order of diffraction would fall in the region of interest.

The grating spectrometer (Oriel MS125) has a focal length of 120 mm and a 75 line/mm diffraction grating blazed at 4.65 µm. The pyroelectric detector is mounted on the outlet side of the spectrometer.
The infrared detector (IR Microsystems μray64) is covered by a CaF$_2$ window (transmission range 0.1-10 μm) and consists of a linear array of 64 pixels. It is well suited to this application for its fast response time, ruggedness, and low cost. Pyroelectric detectors respond to incident radiation, which changes the temperature of the ceramic material of each pixel. The temperature change generates a current that is proportional to the incoming radiance. Employing a chopper at 10 Hz in lock-in technique removes the (out-of-phase) random signal generated by the heat of the detector itself and exposes the pyroelectric array to the required temperature changes to induce a current. The raw output of the array is in number of counts versus pixel position. The former is a measure of radiance and the latter of wavelength position. The long pass filter along with the 75 line/mm grating yield an overall wavelength range of 3.71 μm to 4.99 μm (Section 2.2.2).

2.2.2 Wavelength and Radiance Calibration

Infrared spectra can be obtained by converting the raw output into spectral radiance as a function of wavelength. In order to assign a wavelength position to each of the 64 pixels in the detector array, two narrow band pass filters with centre wavelengths 3.906 μm and 4.594 μm were used in a transmission experiment. This experiment involved recording the number of counts produced by an infrared light source with the narrow band pass filter and without the filter. From the two data points, a linear relationship between pixel number and wavelength was derived. The assumption of linearity was later verified with a third narrow band pass filter (4.041 μm). The difference in the spectral width of one pixel between using two or three filters was 0.2%. The measured wavelength range was from 3.711 μm to 4.987 μm, with a pixel width of 20.3 nm ± 0.2
nm. Repetition of these tests on subsequent days showed that the spectral position of each pixel is accurate to better than 10 nm.

The objective of radiance calibration is to convert the raw signal at every pixel position into spectral radiance \( (\text{W/m}^2/\mu\text{m/sr}) \). An IR-12 Steady State Infrared Emitter (HawkEye Technologies) of emissivity 0.8 was used to relate the raw spectra at four different temperatures between 873K and 1173K, to the corresponding blackbody curves. From this information, calibration factors in terms of \([\text{W/m}^2/\mu\text{m/sr}]/\text{Raw Signal}\) were calculated at every pixel position. The calibration factors are generated by comparing the raw signal to the blackbody monochromatic radiance, \( I_{b,\lambda}(T) \), given by Planck's law (Equation 2-4) in \( \text{W/m}^2/\mu\text{m/sr} \) at the IR source temperature, as seen in Equation 2-3:

\[
\text{Radiance Factor}(\lambda) = \frac{I_{b,\lambda}(T_{\text{source}}) \varepsilon_{\text{source}}}{\text{Raw Signal}(\lambda)}, \tag{2-3}
\]

\[
I_{b,\lambda}(T) = \frac{C_1}{\lambda^5 \left[ \exp \left( \frac{C_2}{\lambda T} \right) - 1 \right]}, \tag{2-4}
\]

where the first and second radiation constants are respectively, \( C_1 = 1.191\times10^{-16} \text{ W·m}^2\cdot\text{sr}^{-1} \) and \( C_2 = 14388 \mu\text{m·K} \) and the emissivity of the IR source is 0.8.

It was observed that the difference between calibration factors decreased as the temperature of the source increased. Since the temperatures in the industrial test were expected to be around 1500K, radiance factors for the temperature retrieval algorithm were linearly extrapolated from the last two calibration sets, 1098K and 1173K (Section 2.2.4). However it was later realized that this IR source introduced significant uncertainty in the analysis of the data from the proof-of-concept test. When a reliable blackbody source is used for radiance calibration the temperature and emissivity results are minimal (less than 1% for isothermal sources). These issues are addressed in Section 2.4.1.
2.2.3 Description of IR Spectra

This section describes the spectral features that appear between 3.7 and 5.0 µm due to the emitted radiation by CO₂ and particles. Some of this radiation is absorbed by room temperature CO₂ present between the combustion gas and the IR sensor (Figure 2-3).

There are three distinct spectral regions that fall in this wavelength interval (Figure 2-2a). From left to right, these regions are characterized primarily by (a) particle-only radiation that is gray due to scattering and the assumed gray nature of the particles between 3.8 and 4.1 µm, (b) a significant decrease in radiation due to absorption by room temperature CO₂, and (c) saturated or blackbody radiation from CO₂ in the boiler. (Note that the use of furnace CO₂ instead of boiler CO₂ in figures and to refer to the 4.56-4.7 µm spectral region, implies that the technique is not restricted to boiler furnaces). Figure 2-2b shows how radiance calibration smoothes some of the jagged instrument features in the raw signal. The spectral features that remain are attributed to the sapphire window present at the proof-of-concept test but not during radiance calibration in the laboratory. Since they were constant for all scans their effect canceled out in the least-squares procedure to retrieve boiler temperatures (Section 2.2.4). The marked decrease in signal on both sides of the spectra is caused by vignetting from the plano-convex lens used to direct the radiation onto the array detector [9].
The analysis of the spectra is aided by the following assumptions: (a) particles and combustion gases are locally isothermal; (b) the CO$_2$ radiation between 4.56 µm and 4.7 µm is saturated over the expected range of temperatures in the boiler; (c) the volume fraction of particles in the line of sight of the IR sensor and the path length of the gas column yield values of the optical depth well above unity in the particle region (i.e., optically-thick medium); and, (d) the scattering due to particles is isotropic.

It is justified to assume that the furnace CO$_2$ region emits like a blackbody. Radiation from polyatomic molecules in the mid-infrared corresponds to changes in a molecule’s vibrational and rotational energy levels. This radiation occurs at discrete wavelengths some of which are so close to one another that radiation appears to be over specific bands of the spectrum. In this work we are concerned not with individual lines but the behaviour of the CO$_2$ band centred around 4.3 µm. Depending on the temperature of the gas-particle stream relative to its surroundings, radiation may be observed as absorbed ($T_{g+p} < T_{\text{surroundings}}$) or emitted ($T_{g+p} > T_{\text{surroundings}}$). The
latter is the case of an industrial boiler where combustion gases are hotter than the surrounding walls (Figure 2-3).

![One-dimensional schematic of the emitted radiation in a boiler.](image)

The amount of light emitted in an industrial boiler through an opening in the wall is described by the Radiative Transfer Equation (RTE). The solution in one-dimension for the case of a gas-particle medium without scattering by particles takes the form [10]

$$I_\lambda(S) = I_\lambda(0)e^{-\tau} + I_{b,\lambda}(1 - e^{-\tau}),$$

(2-5)

where $\tau = \int \beta_\lambda ds$, is the optical thickness of the gas-particle mixture over its path length, and contains the spectral absorption coefficient of the gas and particles and the latter’s scattering coefficient ($\beta_\lambda = \kappa_{g,\lambda} + \kappa_{p,\lambda} + \sigma_{p,\lambda}$). Between 4.56 µm and 4.7 µm, the main sources of emission are high temperature CO$_2$ and particles. This radiation is attenuated by absorption and scattering along the path from 0 to $S$. The radiation from the wall, $I_\lambda(0)$, is likewise attenuated. In the case of a boiler, the temperature of water walls can be roughly half of the combustion gases and therefore the wall radiation term is much smaller than the blackbody radiance $I_{b,\lambda}$ at $T_{g+p}$.

Equation 2-5 predicts that for large values of the optical thickness $\tau = \tau_\lambda$, the radiation leaving the boiler is that of a blackbody (since $e^{-\tau}$ vanishes). The fact that the path length at Nanticoke is
roughly 14 m helps drive the optical thickness up. Also the absorption coefficient of CO$_2$ ($\kappa_g$) in the high temperature portion of the 4.3 $\mu$m band will be significant given its concentration at a height of ten meters above the fire ball ([CO$_2$] $\approx$ 14 mol %) and temperatures around 1500 ±100K. For these conditions (high concentration and temperatures, long path length), the radiation in the 4.56-4.7 $\mu$m range will be that of a blackbody at the gas temperature, i.e., it will be saturated. Figure 2-4 shows how 14% CO$_2$ over a 14 m path length at three different temperatures (1400K, 1500K and 1600K) matches the blackbody lines at those three temperatures in the furnace CO$_2$ spectral region. These results were obtained with RADCAL [11], a one-dimensional solver of the RTE that accounts for absorption and emission by gas and soot but neglects scattering. The saturated property of the CO$_2$ band is the basis of the gas temperature retrieval method described below. Finally, it is worth noting that H$_2$O in the combustion gas will also emit in the furnace CO$_2$ region but not as strongly. For instance, at 4.7 $\mu$m and 1500K, RADCAL shows that the difference in emitted radiance between 14% CO$_2$ and 14% CO$_2$ plus 10% H$_2$O is only 0.01% for the same conditions as in Figure 2-4.
2.2.4 Method of Temperature Measurement

The temperature calculation is based on a least-squared optimization method that compares the theoretical blackbody radiance from Planck’s law, $I_{b,\lambda}(T)$, and the measured raw data after radiance calibration, $I_{\lambda}(T)$. An average over the furnace CO$_2$ spectral region of the regression variable

$$R^2_{g+p} = \sum_{\lambda_{eg+p}} R^2_{\lambda} = \sum_{\lambda_{eg+p}} [I_{b,\lambda}(T) - I_{\lambda}(T)]^2$$

(2-6)

is calculated for each scan. This is done for a series of plausible temperatures (e.g., between 1000K and 2000K). The retrieved temperature ($T_{g+p}$) is that for which $R^2$ is smallest, as shown in Figure 2-5 for one scan with retrieved $T_{g+p} = 1533$K.
2.2.5 Method of Particle Emissivity Measurement

In the particle-only spectral region from 3.8 μm to 4.1 μm where there are no active CO₂ vibrational-rotational bands, the emerging radiation from the boiler is gray. The participating medium consists likely of fly-ash and unburned coal particles whose radius is expected to be between 3 and 50 μm [12]. Radiation will be emitted, absorbed and scattered by these particles. Under the assumption of an optically-thick medium and for isotropic scatterers, Hapke [13] develops an asymptotic solution of the RTE for the radiation emitted by a column of suspended particles following the method of embedded invariance. The premise is that if a thin layer of particles is added to the top of a semi-infinite medium of particles, the emitted radiation from the medium will not change. He calculates five first-order changes caused by the thin layer and then postulates that their sum be zero. The five changes considered are: (a) attenuation by the thin layer of emitted radiance from the semi-infinite medium; (b) emission by the thin layer; (c) scattering by the semi-infinite medium of radiation emitted downwards by the thin layer; (d) scattering by the thin layer of radiation emitted upwards by the semi-infinite medium, and (e) reflection by the semi-infinite layer of radiation first emitted by itself and then scattered.
downwards by the thin layer. The analysis derives an equation for the radiance emerging from the medium as

\[ I_\lambda (T_{\text{medium}}) = H(\mu) \cdot \gamma \cdot I_{b,\lambda} (T_{\text{medium}}). \]  

(2-7)

This expression contains a component of the bidirectional reflectance function called the \( H \) function. For isotropic scatters \( H \) may be approximated as

\[ H(\mu) \approx \frac{1 + 2\mu}{1 + 2\mu\gamma}, \]  

(2-8)

where \( \mu \) is the cosine of the emergence angle of the radiation relative to the normal, and the medium’s albedo factor \( \gamma \) is a function of the particle’s single-scattering albedo \( \omega_p \) according to

\[ \gamma = \sqrt{1 - \omega_p}. \]  

(2-9)

Equation 2-8 differs by less than 4% from the exact values over all \( \mu \) (from 0 to 1) and performs better than that in most places \([13]\). Since the IR sensor is pointed perpendicularly to the body of gas, \( \mu = \cos 0^\circ = 1 \). Then the emitted radiance arriving at the detector can be expressed as

\[ I_\lambda (T_{\text{medium}}) \approx \frac{3\gamma}{1 + 2\gamma} I_{b,\lambda} (T_{\text{medium}}). \]  

(2-10)

Equation 2-10 contains both the effective emissivity of the particle medium, defined as \( \varepsilon_{\text{eff}} = I_\lambda/I_{b,\lambda} \), and, because of the relationship between \( \gamma \) and the particle emissivity \( \varepsilon_p \) (via \( \omega_p \) as explained below), one can derive \( \varepsilon_p \) in the mid-IR.

The relationship between \( \omega_p \) and \( \varepsilon_p \) follows from the definition of the single-scattering albedo of the particles \([13]\)

\[ \omega_p = \frac{\sigma_p}{\kappa_p + \sigma_p}, \]  

(2-11)
which contains the particle’s scattering ($\sigma_p$) and absorption ($\kappa_p$) coefficients. These two quantities may be expressed in terms of the number density of particles $N_p$ and the particle emissivity as follows [6]

$$\kappa_p = \varepsilon_p N_p \pi r_p^2,$$

$$\sigma_p = (1 - \varepsilon_p) N_p \pi r_p^2,$$

where an effective particle radius, $r_p$, has been used instead of a particle size distribution for simplicity. We also assume that diffraction from large particles can be treated as transmission, so that the particle extinction coefficient (the sum of absorption and scattering) is unity, i.e., $\varepsilon_p + (1 - \varepsilon_p) = 1$ [10]. With this information, Equation 2-10 becomes

$$I_\lambda (T_{\text{medium}}) = \frac{3 \sqrt{\varepsilon_p}}{1 + 2 \sqrt{\varepsilon_p}} I_{b,\lambda} (T_{\text{medium}}) = \varepsilon_{\text{eff}} I_{b,\lambda} (T_{\text{medium}}).$$

Another useful variable to define at this point is the brightness temperature of the particles. Their physical temperature ($T_{\text{medium}}$) is assumed to be the same as the gas at a specific location in the field of view of the instrument. The particle brightness temperature ($T_p$) is the temperature of a blackbody that would match the spectral radiance from the gray medium, i.e.,

$$I_\lambda (T_{\text{medium}}) = \varepsilon_{\text{eff}} I_{b,\lambda} (T_{\text{medium}}) = I_{b,\lambda} (T_p),$$

where $T_{\text{medium}} > T_p$. For a blackbody both its physical and brightness temperatures are the same. The particle brightness temperature is retrieved from an analogous regression variable $R_p^2$ (Equation 2-6) with $I_\lambda$ and $I_{b,\lambda}$ from the wavelength range $\lambda_p \in [3.8 - 4.1 \mu m]$.

So far it has been determined that the radiation measured by the IR sensor in the furnace CO$_2$ region will follow a blackbody at $T_{g+p}$ and in the particle region it will be gray. Two temperatures may be retrieved from the spectral data between 3.7 and 5.0 $\mu$m, $T_{g+p} (= T_{\text{medium}})$ and $T_p$, as well
as two emissivities, $\varepsilon_{\text{eff}}$ and $\varepsilon_p$. All these variables may be expressed in a RADCAL simulation, which is useful for the conceptual interpretation of experimental results (Figure 2-6).

![Graph showing simulation of Nanticoke conditions in RADCAL using average values from the proof-of-concept test.](image)

Figure 2-6. Simulation of Nanticoke conditions in RADCAL using average values from the proof-of-concept test.

The slight mismatch between the blackbody curve at $T_p = 1437K$ and the Nanticoke model in the particle region is due to emission by water (assumed = 11 mol %) in the combustion gas. This effect is considered negligible in the rest of the analysis. The room CO$_2$ between the viewing port on the boiler and the IR sensor was modeled as a one-meter layer of air containing 500 ppm of CO$_2$ at 313K.

Finally, from the last equality of Equation 2-15, $\varepsilon_{\text{eff}}$ can be calculated at an appropriate wavelength of the particle region (e.g., $\lambda = 3.95$ µm) as $I_{b,3.95}(T_p)/I_{b,3.95}(T_{g+p})$. Then, Equation 2-14 can be solved for $\varepsilon_p$ as a function of $\varepsilon_{\text{eff}}$:

$$
\varepsilon_p = \left( \frac{\varepsilon_{\text{eff}}}{3 - 2 \varepsilon_{\text{eff}}} \right)^2, \quad (2-16)
$$
which gives the correct result for black particles ($\varepsilon_p = \varepsilon_{\text{eff}} = 1$) and predicts that a medium of gas and gray particles will have an effective emissivity greater than the individual particle emissivity due to in-scattering. The retrieved particle emissivity is a spectral quantity, valid for the same wavelength for which $\varepsilon_{\text{eff}}$ was calculated.

2.3 Results

2.3.1 Industrial Test at a Coal-Fired Boiler (Nanticoke)

A proof-of-concept test with the IR sensor was carried out at Ontario Power Generation’s (OPG) Nanticoke station in February 2006. Nanticoke, located on the north shore of Lake Erie, has eight generating units each consisting of two main cycles: a combustion cycle to produce steam in the coal-fired boiler and a steam cycle to generate electricity in a steam turbine. The station’s total capacity is 3,920 MWe. The experiment took place in the boiler of Unit 4 that fires a mixture of bituminous and sub-bituminous coals. The IR sensor was placed at one of the corner view-ports on level 5 1/2 (elevation over sea level = 203 m), overlooking the fireball from the rows of burners on levels 3 and 3 1/2 (elevation of the latter = 193 m). The instrument was placed in front of the port (Figure 2-7). Safety devices such as a metal conduit and a shield with a sapphire window were necessary to minimize the intense radiant heat from the 38 cm x 25 cm opening.
Figure 2-7. The IR sensor during the industrial test at one of Nanticoke’s coal-fired boilers.

The automatic data acquisition system was set to record one scan every 3 s over a 10 min period, for a total of 200 scans. Although radiation from the water walls (at about 750K) was assumed negligible, the presence of high temperature fly-ash and unburned coal particles added a blackbody-like background to the recorded radiation which was otherwise emitted mostly by CO₂ (Figure 2-6). The CO₂ concentration was estimated by a plant operator to be about 14 mol % (wet) at that level. From the view-port to the far wall inside the boiler there was a distance of 14 m, which was taken as the path length in the RADCAL model (Figure 2-6).

2.3.2 Raw Data Analysis
The analysis of the raw data lent itself to some interesting conclusions, since an overall decrease in the number of counts (a measure of radiance) with time was noticeable. This decrease might be due to either a change in the instrument response function or to a drop in temperature in the boiler. The possibility that CO₂ decreased over time was ruled out since the number of counts decreased evenly throughout the particle and gas spectral regions. By looking at the evolution in
time of two wavelengths—one in the furnace CO_2 emission band (4.60 \mu m) and one in the room CO_2 absorption feature (4.29 \mu m)—it is clear that the instrument response is fairly constant for the absorption band (Figure 2-8), while the furnace CO_2 decreased with time. Thus we discarded the possibility of a change in the instrument response in favour of a slight, yet noticeable drop in boiler temperature.

![Figure 2-8](image)

Figure 2-8. Raw number of counts (a measure of radiance) for two wavelengths (4.60 \mu m from the CO_2 emission band and 4.30 \mu m from the room CO_2 absorption band) versus time.

2.3.3 Temperature and Emissivity Results

Following the methodology outlined in Sections 2.4 and 2.5, \( T_{g+p} \) (physical temperature of the gas and particle medium) and \( T_p \) (brightness temperature of the particles) were calculated for each scan. The results are shown in Figure 2-9.
The average gas temperature was 1533K, which was 96K higher than the average brightness temperature (1437K). These values were used in the RADCAL model of Figure 2-6. The two sets of horizontal lines represent the average temperature in the 0-5 min and 7.5-10 min periods, respectively. For the gas temperature, these values are 1555K and 1479K, which translate in a 4.9% reduction in gas temperature. Using Planck’s function at 4.6 µm (furnace CO₂ region) such a reduction in temperature would be equivalent to an 11.2% decrease in radiance. At the starting time of the test we were advised that the boiler had become stable again after a change in the unit’s electrical load from 440 MWe to 490 MWe. One explanation for the noticeable decrease in temperature may be that the test data captured the transition from the high firing rate required to achieve the new set point in electrical load and the new steady state firing rate to maintain the system at the higher load. It would have been easier to validate this hypothesis with a longer data acquisition period, but unfortunately we were limited in this regard.

The emissivity results appear in Figure 2-10. Panel (a) is the time series data showing moving averages of emissivity and temperature every 45 s. The same emissivity data is then presented as
a function of temperature in (b). The effective emissivity is higher than $\varepsilon_p$ and is less affected by the decrease in temperature over time. The reason why $\varepsilon_{\text{eff}}$ and $\varepsilon_p$ increase with lower temperatures is because $T_p$ remains steadier than $T_{g+p}$ (Figure 2-9). The average emissivity values were $\varepsilon_{\text{eff}} = 0.84$ and $\varepsilon_p = 0.41$ at 3.95 $\mu$m (i.e., at the centre of the particle spectral region).

Figure 2-10. Time series of the particle and effective emissivities for the Nanticoke test with the physical temperature plotted on the secondary axis (a). The same emissivity results are presented versus temperature on (b).

2.4 Discussion
The purpose of this section is to comment on the reasonableness of the results and of the assumptions that support them.

Chui and LeBlanc [14]'s CFD study modeled a very similar boiler to the one where the present test took place. Their temperature distribution 10 m above the top row of burners (i.e., where the IR sensor was located) ranged from 1473K to 1673K. The average temperature retrieved in our test was 1533K and one may conclude that the current results are reasonable.

Linka et al. [2] reported the spectral emissivity of three types of fly-ash particles as a function of temperature. The main components were $\text{SiO}_2$ ($\approx 39$ wt-%), $\text{Al}_2\text{O}_3$ ($\approx 16\text{-}21\%$), $\text{Fe}_2\text{O}_3$ ($\approx 12$-
29% and CaO ($\approx 4-13\%$). Their samples came from German lignite coal that was pulverized to a mean diameter of about 90 $\mu$m. Their definition of emissivity follows Equation 2-1, and is thus a type of effective emissivity. For each wavelength interval they measured normal irradiance from a layer of fly-ash particles and compared it to a blackbody source of known temperature. At 1473K and 3.9 $\mu$m (the closest comparison point with our data) their emissivity values lay between 0.72 and 0.80. Our $\varepsilon_{\text{eff}}$ at 1473K and 3.95 $\mu$m is 0.89 (based on the linear fit of $\varepsilon_p$ with $T$ in Figure 2-10b). Since the emissivity of unburned and devolatilized coal particles is around 0.9 [15] one may infer that significant flame by-pass of coal particles may have occurred in the vicinity of the IR sensor. This issue was identified by Chui and LeBlanc [14] for a similar boiler. Also the higher rank of the coal used at Nanticoke (bituminous and sub-bituminous compared to the lignite samples in [2]) is also known to yield higher emissivities [1] supporting the hypothesis that there may have been unburned char in the field of view of the instrument.

Another comparison may be drawn between the present results and the emissivity measurements from three coal ash samples by Bohnes et al. [3]. Their experimental set up was that of Linka et al. [2]. In [3], at 1473K and 3.9 $\mu$m, $\varepsilon_{\text{eff}} = 0.61, 0.49$ and 0.81, for samples 1, 2 and 3, respectively. The difference is attributed to the high content of Fe$_2$O$_3$ in sample 3 (24.7%) compared to samples 1 (16.2%) and 2 (5.9%). Iron oxides (Fe$_2$O$_3$ and FeO) are known to increase particle emissivity in the 1-5 $\mu$m range due to the strong absorption band of Fe$^{2+}$ around 1 $\mu$m (e.g., [16]). One may conclude that the particles at Nanticoke had significant iron oxide levels.

Perhaps the main difference with [2] and [3] is the trend of emissivity with temperature. In [2], Linka et al. observed an average increase of 0.015 per 100K at 3.9 $\mu$m whereas our data shows a decrease of 0.093 per 100K in $\varepsilon_{\text{eff}}$ (Figure 2-10b). Since the increase in emissivity with
temperature is well established in the literature [4], one plausible explanation is to have recourse to the inverse relationship between emissivity and particle size identified in [4]. In this case, the trend with temperature may be masking the real cause of the decrease, namely an increase in the particle size due to the lower combustion intensity towards the end of the test (when lower values of $T_{g+p}$ were retrieved). An increase in carbon-in-ash level would also correlate with lower temperatures driving up the particle emissivity. However without information about the particles’ size and composition at Nanticoke, it is hard to validate these points, which remain to be explored in future tests.

We will now look at the assumptions that support the present analysis. The assumption of equal temperature between solid particles and combustion gases appears reasonable due to the intense turbulent mixing in the boiler and the location of the IR sensor 10 m above the top row of burners. The furnace CO$_2$ spectral region is easily saturated for the path length at Nanticoke (14 m) and typical CO$_2$ concentrations in the boiler (14 mol %) as shown in Figure 2-5. Using the lower limit of particle volume fractions in pulverized coal applications from [12], $f_v = 3 \times 10^{-5}$, the worst-case particle radius of 50 $\mu$m (in terms of increasing the optical depth), and for a path length of 14 m, $\tau = 6.3$, which can be considered already to fall into the optically-thick medium regime. The assumption of isotropic scattering by particles is common in modeling studies (e.g., [5] and [6]) due to its simplicity, but inaccurate for pulverized coal and fly-ash particles that exhibit forward scattering [17]. However since the current technique is based on a ratio of radiances from wavelength regions one micron apart, the effect of scattering on the retrieved emissivity is probably small. The issue of particle effects on temperature and emissivity has been further developed in [18], which comments on the values of particle size and volume fraction that validate the blackbody assumption in the furnace CO$_2$ region.
2.4.1 Error and Uncertainty Analysis

Due to the prototype nature of the IR sensor two types of error have been distinguished: intrinsic and prototype-bound. Intrinsic errors may be experimental or theoretical whereas prototype errors are only experimental. Prototype errors may easily be reduced through equipment improvements, whereas intrinsic errors are unavoidable with this method. All errors will affect the wavelength and radiance calibrations, the retrieved temperatures and hence, the effective and particle emissivities.

By repeating the wavelength calibration on subsequent days an intrinsic error of ±0.5 pixels, i.e., ±10.1 nm, on the pixel number versus wavelength correlation was observed. Even doubling the estimate of this error to ±20.2 nm did not greatly affect our results. By artificially changing the lowest point in the room CO₂ absorption dip from pixel 36 to 37 in the raw data, the average physical temperature increased from 1533K to 1539K and the particle brightness temperature also changed by six degrees (from 1437K to 1443K). Similarly, by decreasing the position of the CO₂ dip by one pixel, the average gas temperature decreased by five degrees and the brightness temperature by four degrees. Both the effective and particle emissivity changed by less than 0.6% in both scenarios. Thus, from the wavelength calibration one can expect an error less than ±6K in temperature and less than ±0.002 in $\varepsilon_{\text{eff}}$ and $\varepsilon_p$.

The intrinsic random error in the number of counts recorded by the IR sensor is ±2 counts as verified in laboratory experiments. By applying this error randomly to the Nanticoke raw data, the average gas temperature remained unchanged. The change in brightness temperature was only one degree, which will introduce a negligible error in the retrieved emissivities.

The largest prototype error stemmed from the infrared source that was available for the radiance calibration. An uncertainty of $\Delta T = 7$K in the temperature can be expected from the manufacturer’s temperature-versus-voltage information. Since the original radiance calibration
depended on extrapolation from two temperature points \((T_1 = 1173K \text{ and } T_2 = 1098K)\), new radiance factors were generated by maximizing the change in the slope of the linear fit from the baseline case. Two scenarios were considered: a shallower slope based on \([T_1+\Delta T, T_2-\Delta T]\) and a steeper slope from \([T_1-\Delta T, T_2+\Delta T]\). The percent changes were of opposite sign and similar in magnitude. The largest change came from the shallower slope: +9.9% in physical temperature, +7.1% in brightness temperature, -5% in \(\varepsilon_{\text{eff}}\) and -22% in \(\varepsilon_p\). However the steeper slope yielded an increase in \(\varepsilon_p\) of 26% (the other percentage increases were lower in absolute magnitude relative to the shallower slope scenario). No information could be found on the uncertainty of the spectral emissivity of the infrared source.

The IR sensor was recalibrated after the Nanticoke test with a true blackbody source (Mikron M330) and radiance factors (Equation 2-3) were obtained for temperatures between 879K and 1973K [19]. The mean error in retrieving the temperature from spectra by another blackbody source (Boston Electronics Corp. IR-563) between 663 and 1323K was 0.2%. The difference between the two temperatures was well within the 1% uncertainty of the IR-563 temperature. As expected, the discrepancy between the retrieved temperature from the particle and furnace CO\(_2\) spectral regions for the blackbody spectra was 0.4%. The maximum error of 1% in temperature would translate into an error of 1.1% in particle emissivity for the average Nanticoke scan. Thus, it is clear that with a proper radiance calibration the temperature retrieval technique is very robust.

The last intrinsic error is due to misalignment of the radiation source relative to the lens tube of the IR sensor. This error decreases the maximum number of counts that could be expected from a small radiation source, and has been found to be \(\pm10\) counts in laboratory experiments. At Nanticoke the infrared radiation was emitted from a rectangular viewport measuring 38 cm x 25
cm and it is therefore safely assumed that the source of IR radiation filled the field of view of the instrument and that no losses were incurred due to misalignment.

In conclusion, the retrieved variables from the Nanticoke test were influenced mostly by the error in the radiance calibration procedure. Temperatures are accurate to within ±10% (i.e., ±152K), effective emissivities to within ±5% (i.e., ±0.046) and particle emissivities to within ±26% (i.e., ±0.105). By calibrating the IR sensor against a true blackbody reference, temperatures from isothermal sources are accurate to within ±0.2% and particles emissivities to within ±1.1%.

So far in the error analysis it has been assumed that the IR sensor is exposed to radiation from an isothermal source. In a large boiler there will be temperature gradients [14] and the temperature and emissivity values will represent some average of the gas and particle properties in the line of sight of the instrument. In order to estimate the environmental uncertainty that a temperature gradient would introduce in the retrieved variables, the following modeling experiment was conducted in RADCAL: the line of sight of the instrument between the view-port and the middle of the boiler was divided into three equidistant zones (S = 2.3 m) with \( T_{g+p} = 1400\text{K}, 1500\text{K} \) and 1600K. The other variables were taken from the average Nanticoke scan (Figure 2-6). We then computed the spectrum from a medium that combined those three zones in series and retrieved \( T_{g+p} = 1410\text{K} \). This suggests that once CO\(_2\) saturates, it is not affected by the hotter layers behind it and that the retrieved temperature and particle emissivity from the Nanticoke test belong to a zone near the wall of thickness \( S \approx 2.3 \text{ m} \). Since heat transfer modelers are typically interested in the net flux at the wall of a boiler, the current technique will provide an average value of particle emissivity that is representative of that zone.
2.5 Conclusions

The passive, remote approach is a sensible step in the design of combustion diagnostics instrumentation for harsh industrial environments. This chapter described a novel, in-situ technique for measuring particle emissivity in a gas-particle medium where the optically-thick assumption applies. The novelty lies in the use of spectral information in the mid-IR to determine particle emissivity. Advantages of this method include the rugged, portable and inexpensive nature of the IR sensor, with which measurements of particle emissivity at different locations in real boilers can be readily performed. Proof-of-concept of the technique was provided by a successful test in a coal-fired boiler at OPG’s Nanticoke power generation station, where it withstood intense radiant heat and ambient temperatures of more than 60°C. Based on two distinct regions of the emission spectra, the physical temperature of the medium and the brightness temperature of the particles were retrieved. With this information two types of spectral emissivity of particles were calculated at 3.95 µm, $\varepsilon_p$ and $\varepsilon_{eff}$. The former is the physical property to be used in radiation codes where the optically-thick assumption applies. Results in the literature showed that the temperature and particle emissivity from the current test are reasonable. However, the response of the emissivity to variations in temperature was opposite to that found in the literature, although a change in the particle size or composition may be the underlying cause of the observed decrease of $\varepsilon_p$ with temperature. The error and uncertainty analysis section highlighted the need to use a true blackbody source for radiance calibration. With such a source, retrieved temperatures and particle emissivities are very accurate for isothermal systems. The presence of temperature gradients in large industrial furnaces is an environmental uncertainty difficult to determine accurately. A simple model of this uncertainty showed that retrieved temperatures are significantly weighted towards the layer near the wall.
Thus, this technique measures particle emissivities that are particularly applicable to the outer region of a boiler or furnace.

2.6 References


[14] Chui E H and LeBlanc M P 2001 Modeling the performance of a full-scale utility boiler equipped with multiple low NOx burners *Proc. 6th Clean Air Int. Conf. (Porto, Portugal)*


[19] Saari R 2007 A passive infrared sensor for combustion efficiency and process control *MASc Thesis* University of Toronto, Canada

3 An Analytical Expression and Iterative Procedure to Check for Particle Scattering Effects on the Gas Temperature and Particle Emissivity Retrieved by the IR Sensor

3.1 Introduction²
The simplicity of passive, spectroscopy-based sensors for measuring high temperature off-gas properties in real furnaces (Figure 3-1), positions them as appealing alternatives to active sensors (e.g., tunable-diode laser sensors [1]) or extractive sampling techniques [2]. The compromise is that one is limited to the information embedded in the radiation emitted by the off-gas. As an example of the capabilities of a passive process sensor, off-gas temperature and particle emissivity retrieved from a full-scale coal-fired boiler appeared recently in [3].

Figure 3-1. The remote operation of a passive spectroscopic sensor for high-temperature off-gas measurements.

² This chapter is based on [17] which appeared in print in 2008. Some improvements have been made to Section 3.3 (application of Equation 3-26 to a gas-only medium and its spectral validity) over [17].
By measuring the radiance between 4.56 and 4.7 \( \mu \)m emerging from a large column of combustion products and finding the best fit with Planck’s function, one can derive the temperature of the off-gas. This is possible for large optical thickness, since the high temperature CO\(_2\) band at those wavelengths emits as a blackbody. If the off-gas entrains micron-sized particles (e.g., fly-ash or char in [3]) that emit, absorb and scatter radiation, the intensity profile between 4.56 and 4.7 \( \mu \)m may differ from blackbody conditions. Here we explore this possibility and its dependence on gas and particle properties (\( \kappa_g, N_p, r_p \)) by following the method of embedded invariance [4].

Hapke [4] developed the asymptotic solution to the Radiative Transfer Equation (RTE) for a semi-infinite column of suspended particles. By assuming isotropic scattering and a uniform particle size, his analytical approximation for the hemispherical emissivity of a semi-infinite particle layer matches very well the numerical solution by Siegel [5] for particle layer of optical thickness equal to 10, as shown in Figure 3-2.

![Graph showing modeled effective emissivities of an optically-thick particle cloud as a function of the average particle emissivity.](Figure 3-2. Modeled effective emissivities of an optically-thick particle cloud as a function of the average particle emissivity.)
Hapke applied his analytical solution in the development of a radiative-conductive model for planetary infrared (IR) remote sensing [6]: the model allowed him to quantify several phenomena observed in planetary regoliths, such as the solid-state greenhouse effect, thermal beaming, and emittance spectra [7]. Kær et al. [8] spoke of Fluent™ developing an emissivity model for ash deposits based on the method of embedded invariance and cited Hapke [4]. However no information is given as to whether the model will account for the presence of gases in the dust layer and no update is available in the literature at this point.

The aim of Siegel’s numerical solution [5] was to calculate the rate of radiative cooling from a droplet generator system to be used in outer space: the droplets would dissipate waste heat by being in contact with a very low temperature environment. Siegel’s work [5] was mainly concerned with transient effects. The comparison with Hapke (Figure 3-2) is only possible with the special case of an isothermal cloud of particles. Later references to Siegel’s cooling droplet model relate to either transient cooling problems [9,10] or to numerical approximations of radiative transfer in enclosures [11].

Here we apply the method of embedded invariance to a combustion environment, possibly for the first time. We extend Hapke’s analysis by including gas absorption and emission to show in which cases particle effects alter the blackbody profile of an optically thick gas band in the infrared. The final expression depends on $\varepsilon_p$ and $\chi$, which is the ratio of the gas absorption coefficient to the particle extinction coefficient. Our practical motivation was to obtain an approximate analytical solution that could be implemented in the retrieval code of a real-time IR sensor for off-gas temperature and particle emissivity [3]. Analytical solutions offer physical insight and are more readily implemented in real-time retrieval codes than numerical solutions.
(e.g., finite volume or discrete ordinate methods to solve the RTE), which are otherwise more exact.

In the discussion section we comment on the values of $\varepsilon_p$ and $\chi$ for which particle effects should be considered, because they lower appreciably the CO$_2$ radiance profile. In these cases, a simple iterative procedure is used to adjust the temperature and particle emissivity values retrieved from the gas band profile. The whole analysis is applied to data from a coal-fired boiler test [3].

3.2 Effective Emissivity of a Semi-Infinite Medium with Gas and Micron-Sized Particles

By following the method of embedded invariance, Hapke [4] considers five changes to the emerging radiance from a semi-infinite medium of suspended particles after a thin layer of optical depth $\Delta \tau << 1$ and of the same properties, is added. He then postulates that the addition of these changes be zero and calculates the effective emissivity$^3$ of the semi-infinite medium based on an analytical approximation of the bidirectional reflectance $r(\mu_o, \mu)$ for a medium of isotropic scatterers.

It is proposed here that the contribution of a radiation-active gas present in the particle-laden medium will affect all five changes to the emerging radiance. In the following derivation, most variables are spectral.

\footnote{Hapke [4] refers to it as \textit{directional emissivity}.}
3.2.1 Derivation

Assume a semi-infinite column of gas and particles to which a thin layer of the same composition is added. The opaque particles are characterized by their number density \( N_p \), the average cross-sectional area \( A_p \), and the average spectral emissivity \( \varepsilon_p \). Assuming geometric optics \((\lambda < r_p)\), the particle absorption and scattering coefficient can be calculated respectively as \[ \kappa_p = \varepsilon_p N_p A_p, \tag{3-1} \]
\[ \sigma_p = (1 - \varepsilon_p) N_p A_p. \tag{3-2} \]

Then the particle single-scattering albedo \((\omega_p)\) is defined as \[ \omega_p = \frac{\sigma_p}{\sigma_p + \kappa_p} = 1 - \varepsilon_p. \tag{3-3} \]

It is also useful to define the medium’s single-scattering albedo \((\omega_{g+p})\) as
\[ \omega_{g+p} = \frac{\sigma_p + \sigma_g}{\sigma_p + \kappa_p + \sigma_g + \kappa_g} \approx \frac{\sigma_p}{\sigma_p + \kappa_p + \kappa_g} = \frac{(1 - \varepsilon_p) N_p A_p}{N_p A_p + \kappa_g}, \tag{3-4} \]

where it is assumed that scattering due to gas molecules in the infrared is negligible.

The optical thickness of the thin layer is divided into its particle and gas contributions as follows
\[ \Delta \tau = \Delta \tau_g + \Delta \tau_p = \kappa_g \Delta z + (\sigma_p + \kappa_p) \Delta z = \kappa_g \Delta z + N_p A_p \Delta z, \tag{3-5} \]

where the gas absorption coefficient \( \kappa_g \) [m\(^{-1}\)] is the only spectral variable that is needed to define the gas’ effect on the optical thickness of the thin layer. The five changes in the emerging radiance are depicted in Figure 3-3 and will be discussed next.
Figure 3-3. Five changes to the emerging radiance from a semi-infinite gas-particle medium introduced by the addition of a thin (virtual) layer (adapted from [4]).

**Change 1. Attenuation by thin layer of emitted radiance from semi-infinite medium**

Since $\Delta \tau \ll 1$, the exponential term associated with extinction has been expanded in a power series, and only the first-order term in $\Delta \tau$ is kept, i.e., $\exp(-\Delta \tau / \mu) \approx 1 - \Delta \tau / \mu$.

$$\Delta I_1 = I_{\text{with layer}} - I_{\text{without layer}} = \varepsilon_{\text{eff,g+p}}(\mu)I_b \exp\left(-\frac{\Delta \tau}{\mu}\right) - \varepsilon_{\text{eff,g+p}}(\mu)I_b$$

$$\Delta I_1 \approx -\varepsilon_{\text{eff,g+p}}(\mu)I_b \frac{\Delta \tau}{\mu},$$  \hspace{1cm} (3-6)

where $I_b$ is the blackbody radiance at the gas-particle temperature and $\mu$ is the cosine of the emerging angle $e$ (Figure 3-3).

**Change 2. Emission from thin layer**

$$\Delta I_2 = \kappa_g \frac{\Delta \varepsilon_g}{\mu} I_b + \kappa_p \frac{\Delta \varepsilon_p}{\mu} I_b = \frac{\Delta \varepsilon_g}{\mu} I_b + \varepsilon_p N_p \frac{A_p}{\mu} \frac{\Delta \varepsilon_p}{\mu} I_b = \frac{\Delta \varepsilon_g}{\mu} I_b + \varepsilon_p \frac{\Delta \varepsilon_p}{\mu} I_b,$$

$$\hspace{1cm} (3-7)$$
where $\Delta z/\mu$ is the length of the cylinder in Figure 3-3 and it is assumed that particles emit isotropically.

*Change 3.* Scattering by semi-infinite medium into solid angle $\Omega_c$ of radiation emitted downwards from thin layer

The integral over $2\pi$ accounts for all the in-scattering angles $i$:

$$\Delta I_3 = \int_{2\pi} \left( \frac{\Delta \tau_s I_b}{\mu_o} + \varepsilon_p \frac{\Delta \tau_p I_b}{\mu_o} \right) \cdot r(\mu_0, \mu) d\Omega_i, \quad (3-8)$$

where $r(\mu_0, \mu)$ is the medium’s bidirectional reflectance from direction $\mu_0$ into direction $\mu$, $d\Omega_i = -2\pi d\mu_0$, and $\mu_0 = \cos i$.

*Change 4.* In-scattering by thin layer into solid angle $\Omega_c$ of radiation emitted upwards from semi-infinite medium

A fraction $\left( \frac{\varepsilon_{g+p} \Delta \tau}{4\pi \mu} \right)$ of the emitted radiance is scattered by the particles. The integral over $2\pi$ accounts for all the emission angles $e'$:

$$\Delta I_4 = \int_{2\pi} \left( \varepsilon_{\text{eff},g+p} (\mu) I_b \frac{\varepsilon_{g+p} \Delta \tau}{4\pi \mu} \right) d\Omega_{e'}, \quad (3-9)$$

where $d\Omega_{e'} = -2\pi d\mu'$, and $\mu' = \cos e'$.

*Change 5.* Reflection by semi-infinite medium into solid angle $\Omega_c$ of radiation first emitted upwards from itself and then scattered downwards by thin layer
The emitted radiation $\varepsilon_{\text{eff},g+p}(\mu')I_b$ is scattered $\varepsilon_{\text{eff},g+p}(\mu')I_b \frac{\Delta \tau}{4\pi} \frac{\Delta \tau}{\mu_o}$, and then reflected $\varepsilon_{\text{eff},g+p}(\mu')I_b \frac{\omega_{g+p}}{4\pi} \mu_o \Delta \tau r(\mu_o, \mu)$. The double integral accounts for all emission ($\varepsilon'$) and reflection angles ($\iota$):

$$\Delta I_i = \int \int \varepsilon_{\text{eff},g+p}(\mu')I_b \frac{\omega_{g+p}}{4\pi} \frac{\Delta \tau}{\mu_o} r(\mu_o, \mu) d\Omega_i d\Omega'$$

The sum of all changes must equal zero

Since the added layer has the same properties as the semi-infinite medium, we postulate that the sum of the five changes must be zero, i.e., that the addition of the thin layer does not affect the emerging radiance from the semi-infinite medium. Therefore, $\sum_{i=1}^{5} \Delta I_i = 0$. Dividing by $\Delta \tau I_b$ and bringing the first two terms to the RHS and rearranging:

$$\varepsilon_{\text{eff},g+p}(\mu) = \frac{\Delta \tau_g}{\Delta \tau} + \frac{\Delta \tau_p}{\Delta \tau} + \int \left( \frac{\Delta \tau_g}{\Delta \tau} \frac{\mu}{\mu_o} + \frac{\Delta \tau_p}{\Delta \tau} \frac{\mu}{\mu_o} \right) r(\mu_o, \mu) d\Omega_i$$

$$+ \int \left( \varepsilon_{\text{eff},g+p}(\mu') \right) \frac{\omega_{g+p}}{4\pi} d\Omega' + \frac{\omega_{g+p}}{4\pi} \left[ \int \frac{r(\mu_o, \mu)}{2\pi} d\Omega_i \right] \left[ \int \varepsilon_{\text{eff},g+p}(\mu') d\Omega' \right]$$

As stated above, we define

$$\chi = \frac{\Delta \tau_g}{\Delta \tau} = \frac{\kappa_g \Delta z}{\kappa_p + \sigma_p \Delta z} = \frac{\kappa_g}{N_p A_p}$$

In this way, $\frac{\Delta \tau_g}{\Delta \tau} = \frac{\chi}{1 + \chi}$, and $\frac{\Delta \tau_p}{\Delta \tau} = \frac{1}{1 + \chi}$. Then Equation 3-11 becomes

$$\varepsilon_{\text{eff},g+p}(\mu) = \frac{\chi}{1 + \chi} + \frac{\epsilon_p}{1 + \chi} + \frac{\chi + \epsilon_p}{1 + \chi} \int \frac{r(\mu_o, \mu)}{2\pi} d\Omega_i + \frac{\omega_{g+p}}{4\pi} \left[ \int \varepsilon_{\text{eff},g+p}(\mu') d\Omega' \right]$$

$$+ \frac{\omega_{g+p}}{4\pi} \left[ \mu \int \frac{r(\mu_o, \mu)}{2\pi} d\Omega_i \right] \left[ \int \varepsilon_{\text{eff},g+p}(\mu') d\Omega' \right]$$

(3-13)
For a medium of isotropically scattering particles, the bidirectional reflectance $r(\mu_o, \mu)$ is [4]

$$r(\mu_o, \mu) = \frac{\omega_{g+p}}{4\pi} \frac{\mu_o}{\mu_o + \mu} \frac{1}{H(\mu_o)H(\mu)},$$

(3-14)

where $\omega_{g+p}$ is used instead of $\omega_p$ to account for the presence of the gas, and $H(\mu)$ satisfies the integral equation

$$H(\mu) = 1 + \frac{\omega_{g+p}}{2} \mu H(\mu) \frac{1}{\mu_o + \mu} \int_0^{\mu_o} H(\mu_o) \, d\mu_o.$$  

(3-15)

The term $\mu \int \frac{r(\mu_o, \mu)}{\mu_o} \, d\Omega$, which appears twice in Equation 3-13, becomes

$$\mu \int_0^{\mu_o} r(\mu_o, \mu) = \mu \int_0^{\mu_o} r(\mu_o, \mu) \frac{1}{\mu_o} 2\pi \, d\mu_o.$$  

(3-16)

Using Equation 3-14 and then Equation 3-15, we can express Equation 3-16 as

$$\mu \int_0^{\mu_o} r(\mu_o, \mu) \frac{1}{\mu_o} 2\pi \, d\mu_o = \mu \int_0^{\mu_o} \frac{\omega_{g+p}}{2} \frac{H(\mu_o)H(\mu)}{\mu_o + \mu} \, d\mu_o = H(\mu) - 1.$$  

(3-17)

The term $\frac{\omega_{g+p}}{4\pi} \int_0^{\mu_o} \epsilon_{g+p}(\mu') \, d\Omega'$, which also appears twice in Equation 3-13, can be simplified to

$$\frac{\omega_{g+p}}{4\pi} \int_0^{\mu_o} \epsilon_{g+p}(\mu') \frac{1}{2} \epsilon_{g+p}(\mu') \, d\mu'.$$  

(3-18)

With Equations 3-17 and 3-18, Equation 3-13 becomes after some rearranging

$$\epsilon_{g+p}(\mu) = H(\mu) \left[ \frac{\chi + \epsilon_p}{1 + \chi} + \frac{\omega_{g+p}}{2} \epsilon_{g+p}(\mu) \right].$$  

(3-19)

Realizing that the two terms inside the right bracket of Equation 3-19 are constant in $\mu$, we integrate both sides of Equation 3-19 with respect to that variable:
\[ \int_0^1 \varepsilon_{\text{eff},g+p}(\mu) \, d\mu = \left[ \frac{\chi + \varepsilon_p}{1 + \chi} + \frac{\omega_{g+p}}{2} \int_0^1 \varepsilon_{\text{eff},g+p}(\mu') \, d\mu' \right] \cdot \frac{1}{0} \, H(\mu) \, d\mu. \] (3-20)

Since \( \mu \) and \( \mu' \) are dummy variables and the limits are the same, the two integrals with the underbrace are identical. Thus,

\[ \int_0^1 \varepsilon_{\text{eff},g+p}(\mu) \, d\mu = \left[ \frac{\chi + \varepsilon_p}{1 + \chi} \cdot \frac{1}{0} \int_0^1 \varepsilon_{\text{eff},g+p}(\mu') \, d\mu' \right] \cdot 2 \cdot \frac{1}{0} \, H(\mu) \, d\mu \] \quad \text{and} \quad \frac{1}{0} \int_0^1 \varepsilon_{\text{eff},g+p}(\mu') \, d\mu' = \left[ 1 - \frac{\omega_{g+p}}{2} \cdot \frac{1}{0} \, H(\mu) \, d\mu \right]^{-1}. \quad \text{(3-21)}

The integral of \( H(\mu) \) in Equation 3-21 is the first moment of the \( H \) function, \( H_o \) [4]:

\[ H_o = \frac{1}{0} \int_0^1 \varepsilon_{\text{eff},g+p}(\mu) \, d\mu = \frac{2}{1 + \sqrt{1 - \omega_{g+p}}} = \frac{2}{1 + \sqrt{\frac{\chi + \varepsilon_p}{1 + \chi}}} \] \quad \text{(3-22)}

where we used Equations 3-4 and 3-12 to relate \( \omega_{g+p} \) to \( \chi \) and \( \varepsilon_p \). With Equation 3-22, the RHS of Equation 3-21 becomes

\[ \int_0^1 \varepsilon_{\text{eff},g+p}(\mu) \, d\mu = \frac{2}{1 + \sqrt{\frac{\chi + \varepsilon_p}{1 + \chi}}} \cdot \frac{1}{0} \int_0^1 \varepsilon_{\text{eff},g+p}(\mu') \, d\mu' \quad \text{and} \quad \frac{1}{0} \int_0^1 \varepsilon_{\text{eff},g+p}(\mu') \, d\mu' = \left[ 1 - \frac{\omega_{g+p}}{2} \cdot \frac{1}{0} \, H(\mu) \, d\mu \right]^{-1} \] \quad \text{(3-23)}

Inserting Equation 3-23 into Equation 3-19, we can solve for the effective emissivity as a function of \( H(\mu) \)

\[ \varepsilon_{\text{eff},g+p}(\mu) = H(\mu) \left[ \frac{\chi + \varepsilon_p}{1 + \chi} + \frac{\omega_{g+p}}{2} \cdot \frac{1}{0} \int_0^1 \varepsilon_{\text{eff},g+p}(\mu') \, d\mu' \right] = H(\mu) \left[ \frac{\chi + \varepsilon_p}{1 + \chi} \right] \] \quad \text{(3-24)}
For the last step it is useful to realize that \( \omega_{g+p} = 1 - \frac{\chi + \varepsilon_p}{1 + \chi} \), according to Equations 3-4 and 3-12. There are no exact analytical expressions of \( H(\mu) \). For isotropic scatterers, Hapke [4] suggests the following expression, which has an error of less than 4% from the exact numerical solution:

\[
H(\mu) = \frac{1 + 2\mu}{1 + 2\mu \sqrt{1 - \omega_{g+p}}} = \frac{1 + 2\mu}{1 + 2\mu \sqrt{\frac{\chi + \varepsilon_p}{1 + \chi}}}. 
\tag{3-25}
\]

With Equation 3-25, we arrive at the final expression for \( \varepsilon_{\text{eff},g+p}(\mu) \)

\[
\varepsilon_{\text{eff},g+p}(\mu) = \frac{1 + 2\mu}{1 + 2\mu \sqrt{\frac{\chi + \varepsilon_p}{1 + \chi}}} \sqrt{\frac{\chi + \varepsilon_p}{1 + \chi}}. 
\tag{3-26}
\]

### 3.3 Discussion

When there is no gas present, \( \kappa_g = 0, \chi = 0 \) and Equation 3-26 reduces to the effective emissivity of a semi-infinite medium with particles only [3]. The difference between the two is that when a gas is present, instead of \( \varepsilon_p \) the term \( (\chi + \varepsilon_p)/(1 + \chi) \) appears. The case when there are no particles present is less straightforward. The RTE can be solved to get an expression of the spectral radiance emerging from an emitting and absorbing medium at constant temperature:

\[
I(S) = I(0) \exp (-\tau_g) + I_b \left[ 1 - \exp (-\tau_g) \right] 
\tag{3-27}
\]

Thus, the effective emissivity \( \varepsilon_{\text{eff},g} = I(S)/I_b \) is unity, for large optical depths, due to a strong gaseous emitter and/or long path lengths. We can arrive at the same result by taking the limit of Equation 3-26 as \( \chi \) tends to infinity and then applying L’Hôpital’s rule to resolve the indeterminacy:
\[
\lim_{x \to \infty} e_{\text{eff},g+p}(\mu) = \frac{1+2\mu}{1+2\mu\lim_{x \to \infty} \frac{\chi + \epsilon}{1+\chi}} = \frac{1+2\mu}{1+2\mu \cdot 1} = 1.
\] (3-28)

An alternative way of reaching the same conclusion is to replace \((\chi + \epsilon)/(1 + \chi)\) in Equation 3-26 by \(1 - \omega_{g+p}\), as was done already to arrive at Equation 3-24. For a gas-only medium, the particle terms would not exist and one would not neglect the gas scattering coefficient in Equation 3-4, so that the single-scattering albedo would be \(\omega_g = \sigma_g/(\sigma_g + \kappa_g) \ll 1\), since for a strong band \(\kappa_g \gg \sigma_g\). Thus, \(1 - \omega_g \approx 1\) and \(e_{\text{eff},g+p}(\mu) \approx 1\).

So far, we have established that the general equation for the effective emissivity of a semi-infinite medium of gas and particles (Equation 3-26) is able to predict correctly the particle-only and gas-only limits. It is interesting to study the mathematical form of the solution in terms of its main parameters \(\chi\) and \(\epsilon\), as shown in Figure 3-4, which shows the effective emissivity in the normal direction \((\mu = 1)\).

Figure 3-4. Effective emissivity of a semi-infinite medium of gas and particles for some values of \(\chi\) and \(\epsilon\).
Some interesting points may be drawn from Figure 3-4. If there is no gas present ($\chi = 0$), the effective emissivity of the semi-infinite medium is higher than the particle emissivity, with the difference between the two diminishing as the particles approach the blackbody limit. The reason is that the medium’s effective emissivity contains the emission from particles, augmented by the scattering of radiation due to the particles themselves.

Secondly, we consider the effect of gas emission on a purely scattering medium (i.e., $\varepsilon_p = 0$). If there are only particles present ($\chi = 0$), Equation 3-26 predicts correctly that the emerging radiance will be zero since when $\varepsilon_p = 0$ there is no emission due to the particles, regardless of their temperature. However, if we consider the case of a gas-particle medium where $\varepsilon_p = 0$, Equation 3-26 suggests that $\varepsilon_{eff,g+p}(\mu)$ will increase towards blackbody behaviour as the gas component becomes more prevalent, as shown in Figure 3-4. In this case, the gas still emits as a blackbody since $\tau_g >> 1$, but the medium’s effective emissivity is less than unity due to particle scattering.

A third point is the asymptotic behaviour of all the curves as the gas absorption coefficient increases and/or the particle extinction coefficient decreases. For low emissivity particles the single-scattering albedo $\omega_{g+p}$ increases and the otherwise blackbody radiation from a semi-infinite column of gas, will appear gray. This dampening effect caused by particle scattering is diminished as the gas absorption coefficient becomes dominant (i.e., large $\chi$). For $\chi = 1$ and $\varepsilon_p = 0.5$, the effective emissivity is already 5% below unity. We are suggesting that this be the threshold for correcting for particle effects on the spectral profile of saturated gas regions. For $\chi < 1$ and $\varepsilon_p < 0.5$, the reduction in effective emissivity results in appreciable changes on the
secondary variables (e.g., $T_{g+p}$, $\varepsilon_p$) retrieved from the radiance profile, as discussed in Section 3.2.

It is useful to comment on the spectral validity of Equation 3-26. By assuming a semi-infinite medium both in its particle and gas components, i.e., $\tau >> 1$, the method of embedded invariance implies that either $\tau_p$ or $\tau_g$ be $>> 1$. Typically this requires long path lengths, which are common in large combustion systems. Thus, in a gas-particle system that is optically thick in its particle component but optically thin in its gas component over some spectral regions (e.g., when $\kappa_g$ in the wings of an emission band yields a low value of $\tau_g$ such as unity), Equation 3-26 would still be valid.

### 3.3.1 Application to Temperature and Emissivity Data from a Coal-Fired Boiler

It is interesting to study what are the physically meaningful values of $\chi$ that are expected in a specific situation. For the coal-fired boiler in [3] the average spectral absorption coefficient of CO$_2$ at 4.60 µm for typical conditions (14 mol%, 1500K, 1 atm, 14 m path length) can be calculated from the EM2C model based on the statistical narrow band method by Malkmus [14]. This resulted in $\kappa_g = 1.82$ m$^{-1}$. In order to estimate $N_p A_p$, we looked at the range of particle volume fractions in [15] for pulverized coal combustion, which varied between 3x10$^{-5}$ and 2x10$^{-4}$. Then, assuming an average particle radius of either 5 µm or 30 µm, $N_p A_p$ would lay between 0.75 m$^{-1}$ ($f_v = 3\times10^{-5}$, $r_p = 30$ µm) and 36 m$^{-1}$ ($f_v = 2\times10^{-4}$, $r_p = 5$ µm). Therefore, we can expect values of $\chi$ to range from 0.05 ($f_v = 2\times10^{-4}$, $r_p = 5$ µm) to 2.43 ($f_v = 3\times10^{-5}$, $r_p = 30$ µm). From Figure 3-4 we see that the effective emissivity for $\varepsilon_{p,4.60\mu m} = 0.5$ would vary between 0.89 and 0.97 of blackbody radiation (based on the data in [3] for $\varepsilon_{p,3.95\mu m}$ and the observation in [16]
that $\varepsilon_{p,4.60\mu m} \approx \varepsilon_{p,3.95\mu m}$, or slightly larger, for coal ash samples). In this case the temperature (and therefore the particle emissivity data) is not exactly the physical temperature of the gas-particle medium, since the retrieval of $T_{g+p}$ assumed $\varepsilon_{\text{eff},g+p} = 1$ between 4.56 and 4.7 $\mu m$. Since accurate estimates of $\chi$ are difficult to obtain in-situ, the present analysis serves to point out a potential source of uncertainty in the temperature and emissivity calculations that follow the method in [3] without considering particle effects.

3.3.2 Accounting for Particle Effects with an Estimate of $\chi$

If an estimate of $\chi$ is available, an iterative calculation can correct for particle effects in the calculation of temperature and particle emissivity. This procedure is illustrated with the data and methodology in [3]. With $\chi$ and the values for $T_{g+p}$ and $\varepsilon_p$ from a given time, we recalculated $T_{g+p} = T_{g+p,2}$ based on the estimate of $\varepsilon_{\text{eff},g+p}$ (Equation 3-26) and the following equation

$$\varepsilon_{\text{eff},g+p} = \frac{I_b(4.60\mu m, T_{g+p,1})}{I_b(4.60\mu m, T_{g+p,2})}. \tag{3-29}$$

With the new estimate of $T_{g+p,2}$, we updated $\varepsilon_p$ [3] and $\varepsilon_{\text{eff},g+p}$ (Equation 3-26) to be able to calculate $T_{g+p,3}$ from Equation 3-29, noticing that in the second (and subsequent) iteration(s), the numerator is still a function of $T_{g+p,1}$, since that is the retrieved brightness temperature from the 4.56 to 4.7 $\mu m$ region [3]. As stated above, it was also assumed that $\varepsilon_{p,4.60\mu m} \approx \varepsilon_{p,3.95\mu m}$. Convergence was achieved when the difference in $T_{g+p}$ between two iterations was $\leq 1K$. Figure 3-5 shows the results for two sets of values from [3] that differ in the initial value of $\varepsilon_p$. 
Figure 3-5. Temperature (a) and particle emissivity at 3.95 µm (b) for different values of $\chi$ after including particle effects.

If $\varepsilon_{\text{eff},g+p}$ is originally greater than 0.9 (which happened in both cases for $\chi \geq 0.6$), convergence is achieved in at most four iterations. The difference between initial and corrected values of $T_{g+p}$ and $\varepsilon_p$ decreases slightly with greater $\varepsilon_{p,\text{ini}}$ as shown in Figure 3-4. For $\chi = 1.2$ (the mid-point for a pulverized coal boiler as per the calculation in Section 3.3.1), $T_{g+p,\text{final}}$ increased by 3.5% (+52K) for $\varepsilon_{p,\text{ini}} = 0.298$ and by 2.8% (+40K) for $\varepsilon_{p,\text{ini}} = 0.469$. This increase in the final gas-particle temperature resulted in smaller particle emissivities at 3.95 µm, which decreased by 29.9% for $\varepsilon_{p,\text{ini}} = 0.298$ and by 26.7% for $\varepsilon_{p,\text{ini}} = 0.469$. Thus, particle emissivities are more susceptible to particle effects than physical temperatures. With an estimate of $\chi$ for the system under study, one can correct temperature and particle emissivity data.

### 3.4 Conclusions

This chapter complements Hapke’s analysis of the spectral effective emissivity from a semi-infinite particle cloud by adding the effect of gas absorption and emission. The new analytical solution allows one to quantify particle effects (absorption, emission and scattering) on the
blackbody profile that some gas bands display in the infrared under large optical thickness conditions \((\tau_g \gg 1 \text{ and } \tau_p \gg 1)\). We were especially interested in the 4.3 \(\mu m\) emission band of CO\(_2\) at high temperatures but the analysis applies to other spectral regions. The magnitude of particle effects can be gauged from \(\chi\), which is ratio of the gas absorption coefficient to the particle extinction coefficient. For \(\chi < 1\) and \(\varepsilon_p < 0.5\), it is advisable to include particle effects in the calculation of secondary physical properties (e.g., \(T_{g+p}\), \(\varepsilon_p\)) retrieved from the gas band profile, as was demonstrated with the temperature and particle emissivity data from a coal-fired boiler. Correcting for particle effects involves a simple iterative procedure, which was also described.

3.5 References


4 Real Time Gas Temperature and Particle Emissivity at a Basic Oxygen Furnace for Steelmaking

4.1 Introduction

During the decarburization phase of the Basic Oxygen Furnace (BOF) process, oxygen is blown into the molten steel bath to reduce its carbon content from approximately 4% to <0.1% [1]. As a consequence, a column of mostly carbon monoxide (CO) and some carbon dioxide (CO₂) emerges from the mouth of the converter. If the exhaust hood operates at slightly negative pressure, room air will be entrained by the off-gas above the mouth, and a flame will form as air and hot CO mix. The high temperature off-gas entrains solid particles rich in iron oxides and other components from the slag layer, such as CaO, SiO₂, FeO, P₂O₅, Al₂O₃, MnO, and MgO [1]. Measuring the off-gas temperature in real-time at the mouth of a BOF converter presents both challenges and benefits. The harsh environment (high temperatures and particle loadings, constant vibration) precludes the long-term use of thermocouples. Other approaches, such as the application of mid-infrared tunable diode lasers [2] or CO₂ thermometers [3], are still in the technology demonstration stage. The reliability of laser solutions, which require that the laser beam travel across the off-gas column, is hindered by particle scattering and sudden gas temperature differences. Based on the available literature [3,4], commercial CO₂ thermometers rely on a narrow-band model of CO₂ around 4.5 μm at high temperatures ( >873K). They have been used to measure exhaust gas temperatures around 973K in an incinerator. However, their

---

4 This chapter is from a paper submitted for publication [23].
principle of operation is not clearly stated and, to the best of our knowledge, CO₂ thermometers have not been successfully applied in steelmaking furnaces yet. The current proof-of-concept test at a full-scale BOF shows the potential of a recent emission spectroscopy technique [5] to measure gas temperature and particle emissivity in real time.

The benefits of having real-time temperature data are twofold: computational fluid dynamic (CFD) models of the exhaust handling system for a BOF [6] usually resort to estimating off-gas temperature for their boundary condition. Also dynamic temperature data, combined with off-gas concentration, would allow mass and energy balances around the converter, which may serve as the basis for a model-based feedback control strategy. This approach is currently being developed for an Electric Arc Furnace [7] with gas temperature and concentration at the fourth hole, and the same principle may readily be applied to a BOF.

Particle emissivity (εₚ) is a useful variable in CFD codes that include radiation heat transfer sub-models to calculate the radiative source term in the energy conservation equation for fluid flow. For simplicity, while CFD codes normally work with spectrally-averaged particle emissivities [8], the present technique yields spectral emissivity at 3.95 μm. However, for coal ash samples containing Fe₂O₃, MgO, CaO, SiO₂ and SO₃, and for single-component samples of FeO and Fe₂O₃, Bohnes et al. [9] showed that the spectrally-averaged particle emissivity and the emissivity at 3.95 μm are very similar for temperatures above 1200K. All these components are also common to BOF particles [1,10]. The emissivity values of Bohnes et al.[9] and Linka et al.[11] were taken from a layer of particles in a crucible to characterize coal ash deposits on heat transfer surfaces. Thus, they cannot be applied to individual particles entrained in a gas stream.

One of the strengths of the current technique is that it measures εₚ for particles suspended in a high temperature gas column. The emissivity measured by Bohnes et al.[9] and Linka et al.[11] is normally referred to as directional emissivity [12] or effective emissivity (εₑffective) [5] as in the
present study, whereas the individual emissivity ($\varepsilon_p$) is simply called particle emissivity. Both are spectral quantities. The analytical relationship between $\varepsilon_{\text{eff}}$ and $\varepsilon_p$ for semi-infinite particle clouds has been developed by Rego-Barcena et al. [5]

A brief review of the methodology for the retrieval of off-gas temperature and particle emissivity is followed by a description of the experimental set-up and the measuring campaign at a full-scale BOF. The discussion section analyzes the temperature and emissivity values, compares them to the relevant literature, and provides an estimate of the experimental uncertainty.

4.2 Methodology

4.2.1 Temperature and Particle Emissivity Retrieval

The radiation arriving at the infrared (IR) sensor is emitted from the region directly above the mouth of the converter (Section 4.3). At this location, the flame sheet that results from the mixing of CO and atmospheric air, is only starting to form and is neglected. In this way, the radiation source is modeled as a homogeneous column of CO and CO$_2$ with small particles. It has been shown [5] that from the radiance versus wavelength profile between 3.8 and 5.0 $\mu$m, two separate regions may be distinguished for large gas columns with CO, CO$_2$ and particles. The 3.80-4.10 $\mu$m range is characterized by particle-only emission that will appear non-black if the particles are assumed gray in that wavelength window. Between 4.56-4.70 $\mu$m, the radiance arriving at the sensor is a mixture of gas and particle emission. In this region, for long path lengths (e.g., several meters) the radiation is normally saturated, i.e., it follows a black-body curve described by Planck’s distribution in Equation 4-1,

$$I_{b,\lambda}(T) = \frac{C_1}{\lambda^\lambda \left[ \exp(C_2 / \lambda T) - 1 \right]}.$$  \hspace{1cm} (4-1)
where $\lambda$ is the wavelength of the radiation, and the first and second radiation constants are respectively, $C_1 = 1.191 \times 10^{-16}$ W·m$^2$·sr$^{-1}$ and $C_2 = 14388$ µm·K [13]. Since Planck’s distribution is only a function of temperature and wavelength, by fitting the measured intensity profiles in those two wavelength regions to a blackbody profile with temperature as the independent variable, two temperature values may be retrieved [5]: from the gas-particle region, if the condition for saturation is met, the temperature will be some average of the physical temperature of the off-gas ($T_{g+p}$); and from the particle region, a lower temperature ($T_p$) will be measured if the particle emissivity between 3.8-4.1 µm is less than unity. (This temperature is called a brightness temperature to differentiate it from the physical temperature). Figure 4-1 illustrates all these concepts for the conditions at the decarburization phase of a typical heat from the industrial test: an off-gas column diameter of 2.87 m, with a 90% CO + 10% CO$_2$ (by vol.) gas mixture at 1500K and a constant particle emissivity of 0.45 between 3.8-4.1 µm. This scenario is modeled with RADCAL, a 1-D solver of the Radiative Transfer Equation (RTE) that accounts for absorption and emission of gases and particles but neglects particle scattering [14]. The drop in radiance around 4.25 µm is due to absorption by a 4 m air layer at ambient temperature (313K) between the off-gas column and the instrument, containing approximately 300 ppm CO$_2$. The data from the so-called atmospheric CO$_2$ region (atm. CO$_2$ in Figure 4-1) is disregarded in the analysis.
The particle emissivity at 3.95 µm is calculated from the asymptotic solution of the radiative transfer equation (RTE) for semi-infinite gas-particle columns, which depends on the cosine of the emerging angle of the emitted radiation relative to the normal direction $\mu$ (Figure 4-3a below) and the particle emissivity itself (Equation 4-2).

$$\varepsilon_{\text{eff},p}(\mu) = \frac{1 + 2\mu}{[1 + 2\mu \sqrt{\varepsilon_p}]} \sqrt{\varepsilon_p}, \quad (4-2)$$

The effective emissivity of the particle region ($\varepsilon_{\text{eff},p}$) is calculated from the retrieved particle brightness temperature and the temperature of the off-gas [5] as expressed in Equation 4-3

$$\varepsilon_{\text{eff},p} = \frac{I_{b,3.95\mu m}(T_p)}{I_{b,3.95\mu m}(T_{g+p})}, \quad (4-3)$$

where $I_{b,\lambda}(T)$ is Planck’s distribution in Equation 4-1.
The assumption of saturation in the gas-particle region, even for large path lengths, needs to be assessed since particle scattering may lower the otherwise blackbody radiation from CO and CO₂ between 4.56-4.70 µm [15]. If particle scattering effects reduce significantly (> 5%) the blackbody profile between 4.56 and 4.70 µm, Rego-Barcena and Thomson [15] provide an iterative procedure to account for particle scattering. Particle scattering effects are assessed from the initial estimate of particle emissivity and from the \( \chi \) parameter:

\[
\chi = \frac{\kappa_g}{N_p A_p},
\]

which is a ratio of the gas mixture absorption coefficient \( \kappa_g \) to the particle extinction coefficient; the latter is the product of the particle cross-sectional area \( A_p \)—assumed uniform—and the particle number density in the gas column \( N_p \). For \( \chi < 1 \) and \( \varepsilon_p < 0.5 \), it is advisable to correct for particle effects. Thus, estimating \( \chi \) is an essential step towards determining the uncertainty in the measurement of \( \varepsilon_p \) and \( T_{g+p} \). We present next the assumptions that led to the calculation of \( \chi \) for the conditions at the industrial site.

### 4.2.2 Calculating \( \chi \) to Validate the Assumption of Saturation in the Gas-Particle Region

The parameter \( \chi \) is calculated with Equation 4-4 using estimates of \( \kappa_g \), \( N_p \) and \( A_p \). The gas mixture absorption coefficient \( \kappa_g \) at 4.60 µm was determined from the EM2C model based on the statistical narrow band method by Malkmus [16]. Table 4-1 summarizes the simulations for several temperatures and compositions. Since the off-gas at the mouth of the converter is assumed to contain a mixture of CO and CO₂, the values used to calculate \( \chi \) are those in the right-most column in Table 4-1.
Table 4-1. Absorption coefficient $\kappa_g$ [m$^{-1}$] at 4.60 $\mu$m for a 2.87 m gas column at several compositions (vol-%) and temperatures from the EM2C model [16].

<table>
<thead>
<tr>
<th>T [K]</th>
<th>100% CO</th>
<th>100% CO$_2$</th>
<th>90% CO + 10% CO$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1300</td>
<td>0.93</td>
<td>7.39</td>
<td>2.15</td>
</tr>
<tr>
<td>1500</td>
<td>0.84</td>
<td>12.01</td>
<td>2.96</td>
</tr>
<tr>
<td>1700</td>
<td>0.77</td>
<td>16.91</td>
<td>3.85</td>
</tr>
<tr>
<td>1900</td>
<td>0.72</td>
<td>21.55</td>
<td>4.69</td>
</tr>
</tbody>
</table>

Particles from the BOF may be divided into coarse dust and fine dust [17]. Coarse dust, accounting for roughly one third of the total dust by weight [18], has a diameter of between 20 and 1000 $\mu$m in diameter, with approximately 80% (cumulative frequency) of all particle diameters smaller than 500 $\mu$m and 50% below roughly 200 $\mu$m [17]. The other two thirds consist of fine dust. Some researchers [17] reported that 80 wt-% of fine particles displayed diameters less than 100 $\mu$m, and that 50 wt-% were particles with diameters below 30 $\mu$m. However, another study [18] showed that the average particle size should be smaller, and claimed that 91% (cumulative frequency) of fines had diameters less than 2.5 $\mu$m. Ray et al. [19], who did not comment on the presence of coarse particles, reported a diameter range between 0.5 and 15 $\mu$m for a BOF without post-combustion (which is our case). For the purpose of the present study, we will carry out the analysis of particle effects (Section 4.5.4) taking values from the lower estimates of particle diameter as a worst-case scenario. The reason is that for the same mass of particles, it is the smallest size that would have the largest effect on the temperature and particle emissivity retrieved by the IR sensor [15]. Therefore, since Ray et al. [19] found that 89 wt-% of all particles had a diameter between 1 and 10 $\mu$m, 5 $\mu$m is taken here as an average.

The particle shape is assumed to be spherical based on scanning electron microscope images [10,19], so that $A_p = \pi r_p^2$, where $r_p$ is the mean particle radius. The average particle number density $N_p$ for each heat is estimated from the total particle mass and cumulative off-gas volume.
The available process variables from the industrial test were the estimated steel weight at tap and the cumulative oxygen consumption. We approximated the total particle mass \( m_p \) as 1.8\% of the tapped steel \([17,18]\). Then the number of particles \( n_p \) is simply the total particle volume divided by the volume of one particle:

\[
    n_p \approx \frac{m_p}{\rho_p} \frac{4\pi}{3} r_p^3,
\]

where \( \rho_p \) is the particle density \( \approx 5400 \text{ kg/m}^3 \), which is as an average for fine and coarse dust from the data \([17]\)).

The off-gas volume was estimated from the oxygen consumption from a simple overall mole balance around the BOF which assumes that the reaction products are 90\% CO and 10\% CO\_2 according to Reaction 4-6 below. This ratio of CO to CO\_2 is normally used to model the decarburization period \([17]\). In Reaction 4-6 all coefficients have been normalized by the O\_2 coefficient to show the relationship between the number of off-gas moles produced from the reaction of one mol of O\_2 consumed.

\[
    \text{O}_2(\text{g,lance}) + 1.82\text{C}_{(\text{bath})} \rightarrow 0.18\text{CO}_2(\text{g,off-gas}) + 1.64\text{CO}(\text{g,off-gas}) \quad (4-6)
\]

Thus, for each cubic meter of \( \text{O}_2 \) there are 1.82 m\(^3\) of off-gas produced. Since the cumulative oxygen consumption for each heat is reported at 1 atm and 289K, the relationship between oxygen and off gas volumes becomes

\[
    V_{\text{off-gas,289K}} \approx 1.82 V_{\text{O}_2,289K} . \quad (4-7)
\]

This is the same relationship that Chigwedu et al. \([17]\) used in their modeling of the off-gas evolved for a converter assuming a 90\% CO + 10\% CO\_2 mixture. We account for the high temperature of the off-gas at the mouth of the converter by correcting its volume from 289K to \( T_{g+p} \) (the off-gas temperature; for example, 1500K) using the ideal gas law:
\[ V_{\text{off-gas}, T_{p}} = V_{O_{2}, 289K} \frac{T_{\text{exp}}}{289K}. \] (4-8)

Finally, \( N_p \) is just the ratio of the total number of particles to the off-gas volume:

\[ N_p = \frac{n_p}{V_{\text{off-gas}, T_{p}}} . \] (4-9)

The ranges of \( \chi \) for each heat appear in Section 4.5.4.

### 4.3 Experimental

As seen in Figure 4-2, the infrared sensor prototype consists of three main components: standard light-collection optics, a grating spectrometer and a linear-array pyroelectric detector [5]. The optical elements in the lens tube are a 2.5 cm CaF\(_2\) plano-convex focusing lens (focal distance = 75 mm at 588 nm) and a long pass filter (3.60-6.89 \( \mu \)m). The filter blocks incoming radiance between 1.85 and 2.5 \( \mu \)m, whose second order of diffraction would fall in the region of interest.

Wavelength calibration was based on transmission experiments with two narrow band pass filters and yielded an overall wavelength range of 3.52-4.78 \( \mu \)m, as determined immediately prior to the start of the two-day campaign at the industrial site. Radiance calibration was performed with a true blackbody source (Mikron M330) and radiance factors were obtained for temperatures between 879K and 1973K. The data acquisition software was customized to carry out the retrieval algorithms for temperature and particle emissivity in real time. Each acquisition-retrieval cycle lasts a total of 2.025 seconds.
In order to mount the IR sensor to the skirt of the BOF, all components were housed in a heat-resistant NEMA box, fitted with a sapphire window (diameter = 7.5 cm). The box had an intake for instrument air designed so that the air flowed parallel to the outside window surface to prevent particle accumulation on the collection optics. The positive pressure created by the air pressurizing the box kept particles from entering the enclosure. The BOF unit was rated at 168 tonnes. The mouth of the converter had a diameter of 3.04 m. The location of the sensor on the skirt and the view of the inside of the NEMA box are shown in Figures 4-3a and 4-3b, respectively.
Figure 4-3. Schematic of the location of the IR sensor on the BOF skirt (a); and a view of the NEMA box with the IR sensor inside (b). The white box is the grating spectrometer and the metal flange on the side attaches the pyroelectric detector to the outlet of the spectrometer (Figure 4-2).

The field trials took place between the evenings of October 17 and the morning of October 18, 2006 and included seven heats. Three heats had Aim Carbon levels below 0.08% and are referred to as “low carbon heats”. Four had Aim Carbon levels above 0.30% (“high carbon heats”). Other relevant properties about these heats appear in Table 4-2.

<table>
<thead>
<tr>
<th>Heat #</th>
<th>Aim carbon [wt-%]</th>
<th>TD1* carbon [wt-%]</th>
<th>TD1* bath temp. [K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>&gt; 0.30</td>
<td>0.055</td>
<td>1964</td>
</tr>
<tr>
<td>2</td>
<td>&gt; 0.30</td>
<td>0.058</td>
<td>1940</td>
</tr>
<tr>
<td>3</td>
<td>&lt; 0.08</td>
<td>0.037</td>
<td>1999</td>
</tr>
<tr>
<td>4</td>
<td>&lt; 0.08</td>
<td>0.040</td>
<td>1979</td>
</tr>
<tr>
<td>5</td>
<td>&lt; 0.08</td>
<td>0.035</td>
<td>1942</td>
</tr>
<tr>
<td>6</td>
<td>&gt; 0.30</td>
<td>0.078</td>
<td>1990</td>
</tr>
<tr>
<td>7</td>
<td>&gt; 0.30</td>
<td>0.170</td>
<td>1978</td>
</tr>
</tbody>
</table>

TD1* = value at first turn-down.

Table 4-2. Bath properties from the seven heats of the field trials.
4.4 Results

Figure 4-4 shows several sample profiles from the calibrated output of the IR sensor (radiance versus wavelength) before and after the oxygen lance was stopped. The legend refers to the number of seconds before or after the lance went off-line. A qualitative description of this figure may give some insight of the physical processes taking place. In this example, we retrieved the largest temperature from the highest profile (124 s before the oxygen flow ceased). The temperature was slightly lower when the lance stopped and fell considerably afterwards, since there was no more oxygen to generate the off-gas flowing past the IR sensor.

Following the methodology of Rego-Barcena et al. [5], off-gas temperature and particle emissivity at 3.95 µm were retrieved in real-time during a campaign at a 168-tonne BOF converter that spanned seven heats, as shown in Figure 4-5.
Figure 4-5. Off-gas temperature ($T_{og+p}$) and particle emissivity ($\varepsilon_p$) retrieved by the IR sensor for the seven heats during the oxygen-blowing period. The data is reported as a moving average every 4 measurement intervals (8.1 seconds). See text for description of the dashed vertical lines.
The start of the heat is taken as the moment the oxygen lance went on-line. The right-most vertical discontinued line in these plots marks the time when the oxygen was stopped. The other two lightly-dotted vertical lines are the start and end times of the steady state decarburization period (as defined below), from which the average off-gas temperature and particle emissivity values are reported in Table 4-3. As explained in Section 4.5.4, the analysis for particle effects – based on the estimated $\chi$ and the initial (retrieved) value of $\varepsilon_p$ – provided an estimate of the uncertainty in $T_{g+p}$ and $\varepsilon_p$.

We calculated the average temperature and particle emissivity from the decarburization period of all heats, trying to avoid the more pronounced oscillations (see the vertical lines in Figure 4-5 for the start and end times of the so called steady state period of each heat). Within these subsets we observed that the number of emissivity outliers, i.e., values of $\varepsilon_p$ larger than one, became noticeable in heats 6 and 7. The air pressure into the box that housed the IR sensor was increased from 5 psig to 20 psig before heat 6. Since the number of emissivity outliers grew at that point, we decided to stop the field trial. Thus, the magnitude of the temperature and emissivity data for heats 6 and 7 is likely suspect. To remove the effect of outlier points, especially for heats 6 and 7, new average values of $T_{g+p}$ and $\varepsilon_p$ were calculated without the outlier points. This resulted in negligible change in $T_{g+p}$ (<1%) but a significant 10% decrease in $\varepsilon_p$ for heats 6 and 7. Table 4-3 contains the number of data points in the steady state zone of each heat, the average $T_{g+p}$ and $\varepsilon_p$ with all data and without the outliers, as well as the number of emissivity outliers and the percent decrease in $\varepsilon_p$. 
By concentrating on heats 1-5, two groups of particle emissivity data emerge: high carbon heats (1-2) and low carbon heats (3-5). As shown in Table 4-3 the emissivity data for these two groups is significantly different and will be addressed in Section 4.5.2.

### 4.5 Discussion

This section is structured around the two physical properties in the off-gas that were measured \( T_{g+p} \) and \( \varepsilon_p \), and the validation of the assumptions in the retrieval algorithms. It includes also an estimate of the experimental uncertainty in \( T_{g+p} \) and \( \varepsilon_p \) after accounting for particle scattering effects.

#### 4.5.1 Analysis of the Off-Gas Temperature Data

The temperature trace captures well the refining phase in a BOF: soon after the oxygen lance goes on-line, CO and CO\(_2\) are evolved from the liquid bath and flow upwards past the mouth of the converter where the IR sensor is located. The sensor records a steep initial change in the off-gas temperature (e.g., heats 1 and 6; Figure 4-5). In some cases (e.g., heats 3 and 4) there is a sudden rise in temperature before the oxygen lance came on-line, likely from the evolution of warm room air mixed with CO\(_2\)-containing combustion gases from the hot metal charge.
Towards the end of the heat the off-gas temperature drops either because no more oxygen is being supplied (heats 6 and 7; Figure 4-5) or because the carbon content in the bath is very low (heats 1-5). The end result is the same in both scenarios: the evolution of CO stops and there is no more high temperature radiation arriving at the IR sensor, whether from gases or particles entrained by the gas. As a general comparison, the temperatures retrieved from the tunable diode laser by Allendorf et al. [2] from a pilot-scale converter (1 ton, 0.36 m mouth diameter) and a full-scale converter (300 tons, 3.65 m mouth diameter) were, respectively, 1450K and 1800K. The laser sensor was positioned over the mouth of both converters as in our case. Since a laser beam would travel through the full path length of the gas-particle mixture, it is logical to expect higher temperatures from this technique compared to the passive IR sensor, where temperatures and emissivities are weighed toward the outer (and thus, slightly cooler) side of the gas column [5]. In this regard it would be sensible to expect the pilot-scale converter temperatures from the laser technique [2] to be close to the current measurements, as is the case. Further evidence for this hypothesis is provided by comparing the passive IR sensor temperatures with the initial results from a passive CO₂ thermometer (Land Instruments CD1), which is shown in Figure 4-6. The CO₂ thermometer was positioned to look directly at the mouth of the same converter as the IR sensor, but measurements were taken weeks after the present campaign [20]. Since the conditions of the two heats are likely different, Figure 4-6 does not provide a direct validation of the current temperature technique. However, Figure 4-6 does suggest that the temperature from the IR sensor is reasonable.
Another comparison could be made with Nedar [10], who installed a probe 27 m away from the mouth of the converter in a study on particle formation in a BOF. The gas temperature in the middle of the heat (7-15 min) is quoted as 1073-1273K. A decrease of 300-400K in the off-gas temperature between the mouth and the position where particles were collected would seem plausible.

One of the direct applications of the off-gas temperature data is for CFD modelers who would otherwise have to assume an off-gas temperature as a boundary condition for their simulations. For instance, in the design of a secondary exhaust hood for a BOF [6], modelers could have used the present technique to measure the off-gas temperature at the mouth of the converter, to characterize the fumes that needed to be captured by the new exhaust hood.
4.5.2 Analysis of Particle Emissivity Data

BOF particles at the mouth of the converter are a mixture of iron oxides and other flux agents. Once the metal-slag emulsion is formed (6-7 min from the onset of blowing), the main mechanism of solid particle formation is the ejection of metal and slag [10]. From the analysis of 27 heats in a BOF converter, Nedar [10] found that particles were mostly spherical and consisted either of a layer of iron oxide surrounding a solid core of metallic iron, or a porous sphere made of oxidized iron or slag components. The main elements present were Fe, Ca, Mg and Mn. The current technique yields an average particle emissivity that lumps together the contribution of all components, thus simplifying the task of the CFD modeler.

Linka et al. [11] and Bohnes et al. [9] measured spectral effective emissivity from a particle layer of oxides and lignite ash samples with a spectral radiometer in the laboratory. Table 4-4 summarizes their findings and the present data for high- and low-carbon heats (heats 1-5 in Table 4-3) for comparison. Note that effective emissivity [5] applies to a cloud or layer of particles (which describes best the measurements against which we are comparing our data [9,11]). The effective emissivity is itself a function of the individual particle emissivity (Equation 4-2), which is the variable of interest for our study. The effective emissivity of a layer of particles is greater than the particle emissivity because of the in-scattering of radiation which adds to the emission of the particles themselves [5]. It is important to keep in mind that it is the particle emissivity that should be used in a CFD model to describe the radiative properties of particles entrained by a gas. On the other hand, the effective emissivity should be used to characterize the radiative emission emerging from a particle layer, as for example, the emitted radiance from an ash deposit on a heat exchanger surface. In the case of the off-gas of a BOF, the effective emissivity characterizes the radiance leaving a layer of gas and particles and is thus an output of the RTE.
It is interesting that the single-phase components tend to have lower emissivities than the multi-component samples with high iron oxide contents. For example, the emissivity of lignite ash 3, which had the most Fe$_2$O$_3$ of the three (24.7 wt-%), was higher than the emissivity of the single Fe$_2$O$_3$ sample. Since iron(III) oxide has a strong effect on particle emissivity between 1-5 µm due to the strong absorption band of Fe$^{2+}$ around 1 µm [21], we expect BOF particles to have a higher Fe$_2$O$_3$ content than the single-phase and coal ash samples in Bohnes et al. [9]. Instead of having to estimate the average $\varepsilon_p$ from the most-abundant single-phase components, the current technique demonstrates a convenient way to measure in-situ an average particle emissivity for off-gas systems. This aggregated approach seems more accurate given the differences that may exist between single-phase components and mixtures of components.

It was stated above (Table 4-3) that a quantitative difference in $\varepsilon_p$ exists between low- and high-carbon heats. The average particle emissivity for low-carbon heats (2-5) was 0.55 while for high-carbon heats (1-2) it was 0.36. The slightly higher average temperature for high-carbon heats (1517K high-carbon; 1471K low-carbon) likely does not account for the whole difference. Figure 4-7 shows a linear fit of the particle emissivity versus temperature for heats 1 to 5. There is a clear decrease in emissivity with temperature, which was also observed in a coal-fired boiler [5]. However, there remains an apparent distinction between the emissivity of low- and high-carbon heats judging from the vertical location of the lines in Figure 4-7.

<table>
<thead>
<tr>
<th>Component</th>
<th>Low carbon***</th>
<th>High carbon***</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeO$^*$</td>
<td>0.76</td>
<td>0.82</td>
</tr>
<tr>
<td>Fe$_2$O$_3$*</td>
<td>0.59</td>
<td>0.90</td>
</tr>
<tr>
<td>CaO*</td>
<td>0.72</td>
<td>0.62</td>
</tr>
<tr>
<td>SiO$_2$**</td>
<td>0.60</td>
<td>0.48</td>
</tr>
<tr>
<td>MgO**</td>
<td>0.59</td>
<td>0.81</td>
</tr>
<tr>
<td>Lignite ash 1*</td>
<td>0.62</td>
<td></td>
</tr>
<tr>
<td>Lignite ash 2*</td>
<td>0.48</td>
<td></td>
</tr>
<tr>
<td>Lignite ash 3*</td>
<td>0.81</td>
<td></td>
</tr>
</tbody>
</table>

* at 1473K [9]; ** at 1273K [11]; *** present study [Low carbon: heats 3-5, avg. $T = 1471K$; High carbon: heats 1-2, avg. $T = 1517K$; data for $\varepsilon_p$ in Table 4-3 and Equation 4-2 for $\varepsilon_{eff}$.]

Table 4-4. Comparison of $\varepsilon_{eff}$ at 3.95 µm and similar temperatures for relevant oxides [9,11] and present results.
The discrepancy may lie in the different recipes and oxygen lance practices for each type of heat. We looked for a possible explanation in the quantity of additives (lime and dolomite) and the ratio of the estimated steel weight at tap over O$_2$ consumption (Table 4-5). Other factors, such as the initial carbon content of the bath (4.71 wt-%) and the quantity of limestone added (0 kg) were reported to be the same for all five heats.

<table>
<thead>
<tr>
<th>Heat</th>
<th>Aim carbon</th>
<th>Lime [kg]</th>
<th>Dolomite [kg]</th>
<th>Steel/O$_2$ [kg/Nm$^3$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>High</td>
<td>5815</td>
<td>4200</td>
<td>17.10</td>
</tr>
<tr>
<td>2</td>
<td>High</td>
<td>8265</td>
<td>5679</td>
<td>16.27</td>
</tr>
<tr>
<td>3</td>
<td>Low**</td>
<td>3098</td>
<td>4087</td>
<td>17.76</td>
</tr>
<tr>
<td>4</td>
<td>Low</td>
<td>3198</td>
<td>4096</td>
<td>17.69</td>
</tr>
<tr>
<td>5</td>
<td>Low</td>
<td>3134</td>
<td>4101</td>
<td>19.05</td>
</tr>
</tbody>
</table>

* High: Aim carbon > 0.3 wt-%; ** Low: Aim carbon < 0.08 wt-%.

Table 4-5. Additives, estimated mass of steel at tap per O$_2$ consumption for heats 1 to 5.
Table 4-5 shows that high-carbon heats (1 and 2) had more burnt lime (CaO) added and slightly lower steel-O₂ ratio. The amount of burnt dolomite (CaO, MgO) was very similar, although heat 2 had approximately 30% more and it recorded the lowest emissivity. Since the emissivity of MgO is among the lowest in Table 4-4, it could be that this component is playing a more pronounced role in the overall particle emissivity. However, the cause of the observed difference in $\varepsilon_p$ between low- and high-carbon heats is not fully understood at present and remains to be explored in future tests.

While CFD codes normally work with spectrally averaged emissivities $\varepsilon_{\text{tot}}$ (Equation 4-10) to save on computational time, the present technique yields a single spectral emissivity value at 3.95 $\mu$m. However, $\varepsilon_p$ at 3.95 $\mu$m for coal-ash samples agrees closely with the total particle emissivity between 1 and 15 $\mu$m [9], which is a suitable range to calculate $\varepsilon_{\text{tot}}$ since it accounts for 98% of blackbody radiance at 1500K. The major species of the coal samples in that study were Fe₂O₃, MgO, CaO, SiO₂ and SO₃ (wt-% > 5.0), the first four of which are also common in BOF particles [1,10].

\[
\varepsilon_{\text{tot}}(T) = \frac{\int_{\lambda_1}^{\lambda_2} \varepsilon_{p,\lambda}(T)I_{b,\lambda}(T) d\lambda}{\int_{\lambda_1}^{\lambda_2} I_{b,\lambda}(T) d\lambda}.
\]  

But most importantly, the data in Bohnes et al. [9] shows that the relationship $\varepsilon_{\text{tot}} \approx \varepsilon_{3.95\mu m}$ applies to particle layers of the single phases FeO and Fe₂O₃, which are the main components in BOF particles. For example, at 1473K, $\varepsilon_{3.95\mu m} = 0.76$ (FeO) and 0.69 (Fe₂O₃), whereas $\varepsilon_{\text{tot}} = 0.73$ (FeO) and 0.68 (Fe₂O₃). Therefore, we propose that the data in the present study can be used as the total emissivity required by radiation sub-models in CFD codes.
4.5.3 Assumptions in the Retrieval Algorithms

One of the fundamental assumptions of this technique is that the radiation in the 4.56-4.7 µm is saturated (i.e., blackbody radiation). This is easily achievable with enough CO₂ in the gas mixture given a sizeable path length for the gas column (for example, 3 m). We assume that the off-gas at the mouth of the converter was 90% CO + 10% CO₂ by vol., and Figure 4-1 shows that the emitted radiance from the off-gas in the gas-particle wavelength region matches very well the blackbody (BB) line at the same temperature. The question arises then as to what is the minimum CO₂ concentration before we start deviating from saturation conditions. Figure 4-8 addresses this point with modeled radiance profiles for 0, 3, and 8 vol-% CO₂ (balance = CO), all of them at 1500K and 1 atm, with εₚ set to 0.45. The temperatures that would have been retrieved from the gas-particle region appear on the legend of the figure. They are 0.5% lower at worst (0% CO₂ + 100% CO scenario). A similar calculation at lower temperatures showed that from the simulated profile for 100% CO at 900K the retrieved temperature would have been 897K. The reason for the robustness of the temperature retrieval method is the use of absolute radiances in the least-squares minimization algorithm as opposed to the actual shape or slope of the radiance profile [5]. (This discussion assumes that particle scattering effects may be neglected, when in reality they did introduce some uncertainty in Tₑ+ₑ and εₑ; Section 4.5.4).
Finally, one of the assumptions in the asymptotic solution to the RTE [12] is that the particle size parameter $X > 10$ when the medium’s porosity is less than 50%, where $X = 2\pi r_p/\lambda$ and porosity is $(1 - f_v)\%$. From our assumption of $r_{p,avg} = 2.5 \, \mu m$, $X$ varies between 4.0 and 3.4 for $\lambda = 3.95 \, \mu m$ and $\lambda = 4.65 \, \mu m$, respectively. However, the medium’s porosity (as explained in Section 4.5.4) is greater than 99.99%, meaning that particles are so far apart that coherent effects are unlikely and that the emission by particles is isotropic [22].

### 4.5.4 Particle Scattering Effects and Uncertainty Analysis

The scattering of radiance (assumed isotropic) by particles is inversely related to their emissivity, i.e., the lower $\varepsilon_p$ the more light will be scattered. In this case the absolute radiance arriving at the IR sensor would be lower, resulting in lower values of the retrieved temperature from the 4.56-
4.7 μm gas-particle region (which at that point is no longer the physical temperature of the off-gas but the brightness temperature for that wavelength region [15]). The criterion for correcting temperature and particle emissivity data to account for particle effects is based on \( \chi \) and the original value of \( \varepsilon_p \). As a general rule, if \( \chi > 1 \) and \( \varepsilon_p > 0.5 \), particle scattering effects may be neglected [15]. However even if \( \chi \approx 2 \), for \( \varepsilon_p < 0.5 \) it is worth checking the influence of particle scattering on the saturated profile of the gas-particle region as demonstrated below. In Section 4.2.2 we outlined the procedure to estimate \( \chi \) based on the available process variables for the BOF test. Table 4-6 summarizes the process variables that served as inputs, which together with the assumptions in Section 4.2.2, allowed us to find \( \chi \) for each heat. The gas absorption coefficient \( \kappa_g = 2.96 \text{ m}^{-1} \) was taken from Table 4-1 for 90% CO + 10% CO₂ (by vol.) at 1500K. The average particle radius was varied between 2.5 μm and 5 μm since the smallest radius is the worst-case scenario used in this study, and 5 μm represents 89% of all particles in Ray et al. [19]

<table>
<thead>
<tr>
<th>Heat</th>
<th>Expected steel wt. at tap [kg]</th>
<th>( V_{\text{O2,280K}} ) [m³]</th>
<th>( V_0 ) [m³]</th>
<th>( V_{\text{offgas at 1500K}} ) [m³]</th>
<th>( f_r )</th>
<th>( N_p ) [m⁻³] for ( r_p = 2.5 \text{ μm} )</th>
<th>( N_p A_p ) for ( r_p = 2.5 \text{ μm} )</th>
<th>( \chi = \frac{\kappa_g}{N_p A_p} ) for ( r_p = 2.5 \text{ μm} )</th>
<th>( \chi = \frac{\kappa_g}{N_p A_p} ) for ( r_p = 5 \text{ μm} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>138027</td>
<td>8070</td>
<td>0.460</td>
<td>76314</td>
<td>6.0E-06</td>
<td>9.21E+10</td>
<td>1.81</td>
<td>1.64</td>
<td>3.27</td>
</tr>
<tr>
<td>2</td>
<td>137690</td>
<td>8464</td>
<td>0.459</td>
<td>80036</td>
<td>5.7E-06</td>
<td>8.76E+10</td>
<td>1.72</td>
<td>1.72</td>
<td>3.44</td>
</tr>
<tr>
<td>3</td>
<td>142134</td>
<td>8005</td>
<td>0.474</td>
<td>75698</td>
<td>6.3E-06</td>
<td>9.56E+10</td>
<td>1.88</td>
<td>1.58</td>
<td>3.15</td>
</tr>
<tr>
<td>4</td>
<td>141191</td>
<td>7980</td>
<td>0.471</td>
<td>75457</td>
<td>6.2E-06</td>
<td>9.53E+10</td>
<td>1.87</td>
<td>1.58</td>
<td>3.16</td>
</tr>
<tr>
<td>5</td>
<td>142511</td>
<td>7481</td>
<td>0.475</td>
<td>70745</td>
<td>6.7E-06</td>
<td>1.03E+11</td>
<td>2.01</td>
<td>1.47</td>
<td>2.94</td>
</tr>
<tr>
<td>6</td>
<td>140616</td>
<td>7852</td>
<td>0.469</td>
<td>74252</td>
<td>6.3E-06</td>
<td>9.64E+10</td>
<td>1.89</td>
<td>1.56</td>
<td>3.13</td>
</tr>
<tr>
<td>7</td>
<td>140124</td>
<td>7374</td>
<td>0.467</td>
<td>69727</td>
<td>6.7E-06</td>
<td>1.02E+11</td>
<td>2.01</td>
<td>1.47</td>
<td>2.95</td>
</tr>
</tbody>
</table>

Table 4-6. Variables used in the calculation of \( \chi \) at \( r_p = 2.5 \text{ μm} \) and 5 μm to check for particle scattering effects on \( T_{g+p} \) and \( \varepsilon_p \).

The iterative procedure to adjust \( \varepsilon_p \) and \( T_{g+p} \) has been outlined elsewhere [15]. We will look first at the worst-case scenario (\( r_p = 2.5 \text{ μm} \), lowest \( \chi \)) for both low- and high-carbon heats in Table 4-3 (above): heat 1 (high-carbon; \( \chi = 1.64 \)) and heat 5 (low-carbon; \( \chi = 1.47 \)). The average \( \varepsilon_p \) for
the steady state zone were 0.368 (heat 1) and 0.557 (heat 5). Similarly, the retrieved temperatures from the gas-particle region were 1527K for heat 1 and 1473K for heat 5 (Table 4-3). The corrected values of $\varepsilon_p$ decreased to 0.286 (-22.3%; heat 1) and 0.447 (-19.8%; heat 5), while the new values of the off-gas temperature increased to 1564K (+2.4%; heat 1) and 1500K (+1.8%; heat 5). For the average $r_p = 5 \mu m$, the changes are more modest. After taking particle scattering effects into account, particle emissivities were 0.324 (-12.0%; heat 1) and 0.499 (-10.4%; heat 5), while the off-gas temperatures rose to 1546K (+1.24%; heat 1) and 1486K (+0.88%; heat 5). Thus, the uncertainty in the retrieved variables (which hinges on the assumption of an average $r_p$ for BOF particles) is at most -22% for $\varepsilon_p$ and +2.4% for $T_{g+p}$. Low- and high-carbon heats display similar uncertainties, which decreased by approximately 50% when the assumed particle radius doubled to 5 $\mu m$. Since normally the particle size distribution at an industrial furnace is not measured in real-time –from which one may determine an average $r_p$–, the nominal maximum uncertainty of the IR sensor for $\varepsilon_p$ around 0.4 is -20% and for temperatures around 1500K is +2%. This uncertainty should be born in mind when considering the data in Table 4-3 and Figure 4-5.

A smaller source of uncertainty in the retrieved variables stems from the radiance calibration procedure. To estimate it, we compared the retrieved and actual temperatures from the radiation of a blackbody source (Boston Electronics Corp. IR-563), which was different from the one used for radiance calibration (Mikron M330; Section 4.3). The mean error was 0.2% between 663K and 1323K, which is well within the 1% temperature uncertainty of the IR-563 source. As expected, the discrepancy between the retrieved temperature from the particle-only and gas-particle spectral regions for the IR-563 blackbody spectra was very small (0.4%).
4.6 Conclusions

This chapter summarizes the successful implementation of a novel IR sensor in a steelmaking furnace to measure gas temperature ($T_{g+p}$) and spectral particle emissivity ($\varepsilon_p$) based on mid-infrared emission spectroscopy. The major conclusions from the study are as follows:

1. Real-time (every 2 s) profiles of $T_{g+p}$ and $\varepsilon_p$ signals were presented for all the heats and average values from the decarburization zone were calculated (Table 4-3).

2. The estimated uncertainty in the measurements was dominated by particle scattering effects and was at worst 2% in $T_{g+p}$ around 1500K and 20% in $\varepsilon_p$ between 0.2 and 0.8. Other sources of uncertainty in $T_{g+p}$ were the radiance calibration procedure (0.2%) and the assumption of 10 vol-% CO$_2$ in the off-gas column (0.5%).

3. Off-gas temperatures from the present study compare favorably with those from published in-situ tests with a tunable-diode laser and a CO$_2$ thermometer.

4. Previous studies measured the emissivity from a layer of particles, and could not provide an average individual particle emissivity that may be used in CFD codes to model radiation from particles in a gas. This observation renders the current particle emissivity data unique.

5. The decreasing trend between $\varepsilon_p$ and temperature agrees well with trends observed in a coal-fired boiler with the same IR sensor.

6. The average emissivity from the decarburization phase of low-carbon heats was noticeably higher than that of high-carbon heats (0.55 and 0.36, respectively), and although temperature plays a role, this difference is not fully understood at present.

7. This technique may be applicable to other metallurgical batch processes that meet the following criteria: presence of a high temperature gas stream ($T > 1000$K) that entrains dust particles of average diameters greater than 5 $\mu$m, and with enough CO/CO$_2$ and path length (for
example, a 2 m off-gas column with 10 vol-% CO₂ or a mixture of CO/CO₂). These conditions may apply to electric arc furnaces, cement kilns, and coal-fired boilers.

8. Real-time off-gas temperature and particle emissivity data will be likely useful to CFD modelers for setting temperature boundary conditions, and to process control professionals who would like to perform continuous energy balances around the furnace.

4.7 References


[18] U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards 1995
*AP-42: Compilation of air pollutant emission factors, Volume 1: Stationary point and area sources* (5th ed., Research Triangle Park, NC, section 12.5)


[22] Hapke B 2007 University of Pittsburgh, Pittsburgh, Pennsylvania, personal communication

5 Process Control Implications of Measuring Real-Time Gas Temperature and Particle Emissivity in a Basic Oxygen Furnace

5.1 Introduction

The Basic Oxygen Furnace (BOF) is an energy, resource and capital intensive facility, where slight productivity improvements translate into a significant competitive advantage within the current global steel market. A simple tool for improvement is the accurate and reliable end-point prediction for the oxygen lance in top-blown furnaces. Productivity gains – and a whole range of environmental and energy benefits – are realized by avoiding costly reblows and trimming oxygen injection to its necessary minimum to meet carbon, temperature and phosphorus targets. End-point strategies rely on static material and energy balance models, and on real-time temperature and composition data near the end of the heat [1]. Static models are not entirely reliable due to the uncertainty of the input parameters and the complexity of simulating accurately the refining reactions. Skilled operators make up for the unreliability of charge models, but they too can be helped with real-time information towards the end of the heat. Real-time bath data is typically obtained with a sublance [2,3] but this practice is not dominant, especially among older converters, due the high retrofitting cost. Another less researched source of real-time information is to correlate the off-gas properties with the temperature and composition of the steel bath. Allendorf et al. [4] shone a mid-infrared tunable diode laser (TDL) across the mouth of a lab-scale and a full-scale converter in order to measure the temperature and

5 This chapter is from a paper submitted for publication [14].
the ratio of CO to CO₂ in the off-gas. However, laser sensors have still to be adopted by industry on a consistent basis. Active sensors, like TDL lasers, in the harsh environmental condition of a BOF face problems such as vibration, beam alignment and beam steering, and –probably the greatest challenge– changing beam attenuation due to transient particle loadings in the off-gas. A second diagnostic technique relevant to BOFs is extractive sampling of the off-gas downstream of the exhaust duct. This concept has been implemented in the Electric Arc Furnace (EAF) where measurement of the off-gas composition (CO, CO₂, H₂ and O₂) at the fourth hole has enabled many shops to improve their productivity and optimize the use of resources [5]. For example, after a typical installation of an extractive gas analyzer to control process inputs at Tamsa (Mexico), the plant realized significant reductions in the consumption of electrical energy (4.3%), natural gas (42.3%), total carbon (10.4%) and oxygen (3.3%) [6]. This technology has the potential of being equally applicable to a BOF converter. A third diagnostic technique to obtain information from the off-gas is passive (or remote) optical sensors. These sensors work by analyzing the visible or infrared radiation from the off-gas. Their advantage lies in being rugged (few or no moving parts), remote and fast (for example, one measurement per second). An example of a visible optical sensor, which works well for low carbon heats in full-combustion furnaces, has been described by Sharan [7]. This device captures the reduction in flame radiance when the off-gas flow rate decreases sharply because the bath has reached the steady-state minimum carbon concentration (here referred to as minimum carbon). From this information the off-gas visible radiance is correlated with the carbon concentration in the steel. The visible light sensor is unsuitable for converters with suppressed combustion or with post-combustion, since the sensor collects the visible radiance of the flame. The optical sensor used in this study [8,9] is similar to that described by Sharan [7] but has the advantage of working well under suppressed combustion conditions since it analyzes the mid-infrared emission of CO and CO₂, whether there
is a flame or not. This is due to the fact that both CO and CO$_2$ at high temperatures have emission bands in the mid-infrared, for example between 4 and 5 µm, but not at visible wavelengths (400 to 700 nm).

The purpose of these measurements is to provide BOF operators –especially those of converters without a sub lance or operating under suppressed combustion– with advance notice of minimum carbon. The drop in flame or gaseous radiance that characterizes minimum carbon and some advance notice –from the shape of the particle emissivity signal– may be possible with the current optical sensor. This device measures the off-gas temperature and the emissivity of the particles entrained by the off-gas in real time (every two seconds). In order to determine the usefulness of the new sensor as a diagnostic tool, the first turndown bath parameters (temperature, carbon, and phosphorus) have been correlated with changes in the off-gas temperature and particle emissivity. A new variable, called minimum carbon time, is defined as the time between the observed peak in particle emissivity and the end of the oxygen blow, and is used to interpret the first turndown data. Based on the findings for low carbon heats, a possible end-point strategy for oxygen injection is proposed.

5.2 Data Acquisition and Analysis

Two sources of data were analyzed. The off-gas temperature and emissivity of particles entrained by the off-gas were measured in real-time with a mid-IR sensor; and the process data (aim and first turndown concentrations for carbon and phosphorus, bath temperature, as well as the oxygen lance status, height and oxygen flow rate in real-time) were provided by U.S. Steel Canada (formerly Stelco Inc.-Hamilton Works).
5.2.1 Data from the IR sensor

The IR sensor and retrieval algorithms and a proof-of-concept test in a coal-fired boiler have been documented elsewhere [8,10]. The specifics of the current field trials—from the point of view of the measurement methodology—were described previously [9]. This chapter presents the process control implications of the off-gas temperature and particle emissivity data when combined with the bath properties at first turndown.

The IR sensor consists of collection optics, a grating spectrometer and a 64 pixel pyroelectric array (Figure 5-1a). It is calibrated to measure radiance [W/m²/µm/sr] versus wavelength [µm] between 3.52 and 4.78 µm. During the test at the 168 tonne BOF, the sensor was housed in a heat resistant (NEMA) enclosure bolted to the skirt of the furnace, just above the mouth of the converter (Figure 5-1b).

The retrieval analysis assumes a gas and particle column that is optically thick and where micron-sized particles scatter isotropically. This is the case of the 3 m long off-gas column in the...
BOF containing mostly CO and some CO$_2$, as well as particles [9]. These conditions justify that (a) the measured radiance profile between 3.80 and 4.10 µm will be that of a gray body when the particle emissivity is less than unity, and (b) that the radiance profile in the gas-particle region (4.56-4.70 µm) will be that of a black-body at the temperature of the off-gas, provided that particle scattering effects may be neglected. Through a least-squares minimization procedure and using Planck’s equation for a blackbody (Equation 5-1) – which depends only on temperature at a given wavelength – two temperatures are retrieved from the particle-only and gas-particle wavelength regions: $T_p$, called the particle brightness temperature, and $T_{g+p}$, the physical temperature of the off-gas, respectively.

\[
I_{b,\lambda}(T) = \frac{C_1}{\lambda^5\left[\exp\left(\frac{C_2}{\lambda T}\right) - 1\right]}, \tag{5-1}
\]

where $\lambda$ is the wavelength of the radiation, and the first and second radiation constants are respectively, $C_1 = 1.191E-16$ W·m$^2$·sr$^{-1}$ and $C_2 = 14388$ µm·K [11]. The particle emissivity ($\varepsilon_p$) at 3.95 µm is calculated from the asymptotic solution of the radiative transfer equation (RTE) for semi-infinite gas-particle columns, which depends on the cosine of the emerging angle of the emitted radiation relative to the normal direction $\mu$ (Figure 5-1b) and the particle emissivity itself (Equation 5-2).

\[
\varepsilon_{\text{eff},p}(\mu) = \frac{1+2\mu}{\left[1+2\mu\sqrt{\varepsilon_p}\right] \sqrt{\varepsilon_p}}, \tag{5-2}
\]

The effective emissivity of the particle region ($\varepsilon_{\text{eff},p}$) is calculated from the retrieved particle brightness temperature and the temperature of the off-gas [8] as expressed in Equation 5-3

\[
\varepsilon_{\text{eff},p} = \frac{I_{b,3.95\mu m}(T_p)}{I_{b,3.95\mu m}(T_{g+p})}, \tag{5-3}
\]

where $I_{b,\lambda}(T)$ is Planck’s distribution in Equation 5-1.
The assumption of true blackbody radiance in the gas-particle region, even for large path lengths, needs to be assessed since particle scattering may lower the otherwise blackbody radiation from CO and CO$_2$ between 4.56-4.70 µm [10]. If particle scattering effects reduce significantly (> 5%) the blackbody profile between 4.56 and 4.70 µm, Rego-Barcena and Thomson [10] provide an iterative procedure to correct the temperature and particle emissivity values. It was estimated that the uncertainty due to particle scattering in the magnitude of the off-gas temperature and particle emissivity was at worst 2% in $T_{g+p}$ around 1250 °C and 20% in $\varepsilon_p$ between 0.2 and 0.8 for the field trials at U.S. Steel Canada [9].

5.2.2 Process Data: Oxygen Lance and Bath Properties
The process data from U.S. Steel Canada for eight heats is shown in Table 5-1. Up-to-the-second information about the status of the oxygen lance, its height and oxygen flow rate for each heat was also provided. The heats were grouped based on their aim carbon contents. Heats 1, 2, 6 and 7 had aim carbon levels greater than 0.30 and are labeled “high-carbon heats”. The aim carbon for heats 3, 4, 5 and 8 was smaller than 0.08 and they are referred to as “low-carbon”. The variable $T_{Diff}$ represents the difference between the first turn down temperature and the aim temperature of the bath.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.325</td>
<td>0.055</td>
<td>1582</td>
<td>1193</td>
<td>0.75</td>
<td>1691</td>
<td>109</td>
<td>0.015</td>
<td>0.049</td>
<td>0.009</td>
<td>81.6%</td>
</tr>
<tr>
<td>2</td>
<td>0.325</td>
<td>0.058</td>
<td>1582</td>
<td>1177</td>
<td>0.74</td>
<td>1667</td>
<td>85</td>
<td>0.015</td>
<td>0.051</td>
<td>0.004</td>
<td>92.2%</td>
</tr>
<tr>
<td>3</td>
<td>0.075</td>
<td>0.037</td>
<td>1607</td>
<td>1387</td>
<td>0.86</td>
<td>1726</td>
<td>119</td>
<td>0.070</td>
<td>0.045</td>
<td>0.007</td>
<td>84.4%</td>
</tr>
<tr>
<td>4</td>
<td>0.075</td>
<td>0.040</td>
<td>1599</td>
<td>1353</td>
<td>0.85</td>
<td>1706</td>
<td>107</td>
<td>0.070</td>
<td>0.042</td>
<td>0.005</td>
<td>88.1%</td>
</tr>
<tr>
<td>5</td>
<td>0.050</td>
<td>0.035</td>
<td>1610</td>
<td>1283</td>
<td>0.80</td>
<td>1669</td>
<td>59</td>
<td>0.015</td>
<td>0.041</td>
<td>0.007</td>
<td>82.9%</td>
</tr>
<tr>
<td>6</td>
<td>0.595</td>
<td>0.078</td>
<td>1555</td>
<td>1343</td>
<td>0.86</td>
<td>1717</td>
<td>162</td>
<td>0.015</td>
<td>0.048</td>
<td>0.019</td>
<td>60.4%</td>
</tr>
<tr>
<td>7</td>
<td>0.595</td>
<td>0.170</td>
<td>1555</td>
<td>1321</td>
<td>0.85</td>
<td>1705</td>
<td>150</td>
<td>0.015</td>
<td>0.045</td>
<td>0.019</td>
<td>57.8%</td>
</tr>
<tr>
<td>8</td>
<td>0.075</td>
<td>0.037</td>
<td>1607</td>
<td>1235</td>
<td>0.77</td>
<td>1733</td>
<td>126</td>
<td>0.070</td>
<td>0.048</td>
<td>0.005</td>
<td>89.6%</td>
</tr>
</tbody>
</table>

$T_{Diff} = TD1 T - Aim T$; **P% reduct. = (In P - TD1 P)/In P %.

Table 5-1. Aim, first turn down (TD1), hot metal (HM) and initial (In) process data for the eight heats. Compositions are expressed in wt-%.
Figure 5-2. Off-gas temperature ($T_{g+p}$; dotted line), particle brightness temperature ($T_p$; solid gray line), particle emissivity ($\varepsilon_p$; solid black line), end of oxygen blow line (solid vertical line) and minimum carbon time ($\tau_{MC} \equiv$ peak emissivity time - end of oxygen time; see the two vertical lines in each plot) for eight heats at a 168 tonne BOF converter. Data is time-averaged every 6 cycles or 12.2 s.
Figure 5-2 shows the evolution of the two temperature variables \((T_p, T_{g+p})\) and the particle emissivity \((\varepsilon_p)\) from the IR sensor. To smooth out the original data, which was acquired every 2.025 s, Figure 5-2 uses a moving average every six data points. The moment at which the oxygen lance went off-line is marked by a solid vertical line reaching from top to bottom. The time near the end of the oxygen blow when the particle emissivity peaked—and the particle temperature started to drop—is also identified by a vertical, thin line.

The time elapsed between those two vertical lines is a new variable introduced here, which is called *minimum carbon time* \((\tau_{MC})\) and is reported in seconds for every heat in Figure 5-2. This variable is set to zero when the oxygen lance went off-line before \(\varepsilon_p\) peaked or \(T_p\) started to drop, which happened in heats 6 and 7, and was about to happen in heat 5 (the lowest \(\tau_{MC}\) recorded, 11 s). The off-gas temperature also dropped before the oxygen injection stopped (heats 1-5, 8) but its decrease was less sharp than that of the particle brightness temperature. In terms of finding a suitable marker to signal operators of an important process change in the bath, the combination of a drop in \(T_p\) (instead of \(T_{g+p}\)) and the \(\varepsilon_p\) peak seems a reasonable choice. The name *minimum carbon* comes from the realization that when \(\tau_{MC} > 0\), \(TD1\ C\) had reached a bulk steady minimum value (between 0.035 and 0.058 wt-%) regardless of how long the oxygen blow was continued (Section 5-3). When \(\tau_{MC} = 0\), \(TD1\ C\) was significantly higher, 0.078 in heat 6 and 0.170 in heat 7.

Figure 5-3 shows the correlation of the newly defined variable \((\tau_{MC})\) with the first turn down bath properties. We have also included the aim values for the four low carbon heats. These plots will provide some insight into the final stage of the decarburization process.
The time at which the particle emissivity peaked (near the end of the heat) relative to the duration of the oxygen blow is shown in Table 5-2. Soon before this peak in emissivity, the operator should have seen an increase in the radiance of the flame. This is discussed in the following section.
Table 5-2. Time at which $\epsilon_p$ peaked expressed as a percentage of the total oxygen injection time.

<table>
<thead>
<tr>
<th>Heat 1</th>
<th>Heat 2</th>
<th>Heat 3</th>
<th>Heat 4</th>
<th>Heat 5</th>
<th>Heat 6</th>
<th>Heat 7</th>
<th>Heat 8</th>
</tr>
</thead>
<tbody>
<tr>
<td>87%</td>
<td>86%</td>
<td>90%</td>
<td>94%</td>
<td>99%</td>
<td>100%</td>
<td>100%</td>
<td>86%</td>
</tr>
</tbody>
</table>

Finally, a clarification regarding the absolute value of the temperature and emissivity data in Figure 5-2. The absolute value of the temperature and particle emissivity values for heats 6 through 8 was likely compromised after the air pressure into the enclosure of the IR sensor was raised before heat 6 started [9]. However, to define $\tau_{MC}$ accurately all that is necessary is to track the relative change in these variables in order to observe the drop in $T_p$ and the peak in $\epsilon_p$. These features were consistently found in all heats.

5.3 Discussion

5.3.1 Trends in the Correlation between Bath Properties and $\tau_{MC}$

The temperature trace in Figure 5-2 captures well the refining phase in a BOF: soon after the oxygen lance goes on-line, CO and CO$_2$ evolve from the liquid bath and flow upwards past the mouth of the converter where the IR sensor is located. The sensor records a steep initial change in the off-gas temperature (for example, heats 1 and 6). In some cases, such as in heats 3 and 4, there is a sudden rise in temperature before the oxygen lance goes on-line, likely from the evolution of warm room air mixed with CO$_2$-containing combustion gases from the hot metal charge. Towards the end of the heat the off-gas temperature drops either because no more oxygen is being supplied (heats 6 and 7, the only ones with $\tau_{MC} = 0$) or because the carbon content in the bath is already close to its steady-state minimum value (heats 1-5, 8; Figure 5-3a). The end result is the same in both scenarios: the evolution of CO is reduced significantly and
there is no more high-temperature radiation arriving at the IR sensor, whether from gases or particles entrained by the gas.

For heats with $\tau_{MC} > 0$ (1-5, 8), the particle emissivity peaked around 90% of the oxygen injection time and then decreased sharply (Figure 5-2 and Table 5-2). This observation agrees well with Sharan’s findings with a visible light sensor [7]. It is after 80-90% of the total oxygen blown that the decarburization reaction between dissolved carbon in the liquid metal and gaseous oxygen, switches control regimes: from being controlled by the oxygen supply (gas diffusion control) to being limited by the carbon concentration in the liquid phase (mass transfer control) [12,13]. The transition in the decarburization mechanism is normally estimated to take place at carbon levels of about 0.1% to 0.3%. At this point there is a sharp decrease in the evolution of CO, which lowers the off-gas emitted radiance. Carbon concentrations quickly stabilize around 0.04% [7], as can be observed in Figure 5-3a. Further oxygen injection also results in the (exothermic) oxidation of molten iron to form FeO, which travels to the slag layer while raising the bath temperature. This last feature—an increase in bath temperature with continued oxygen injection— is well exemplified by the low carbon heats in Figure 5-3b. The rise in temperature levels off from about 29 °C/min $O_2$ injection for $\tau_{MC} < 130$ s, to 3 °C/min $O_2$ injection for $\tau_{MC}$ between 130 s and 260 s. (It should be noted that because Figure 5-3 consists of data for similar yet individual heats, these temperature increase rates should only be taken as part of a qualitative explanation of what will likely happen with prolonged oxygen injection. To be quantitative one would need to track the temperature increase of the same heat at different oxygen injection times.) In all heats the targets for carbon and temperature were achieved, even though Figure 5-3 shows only the aim values of the four low carbon heats. In order to reach their carbon target (see bars in Figure 5-3a), coke and other carbon-containing alloy additives such as manganese or chromium are introduced at tap. The phosphorus values were lower for the six heats with $\tau_{MC} >$
0, which met their target (Figure 5-3b). However in heats 6 and 7, \( \tau_{MC} = 0 \) and the first turndown phosphorus (0.019% in both cases) was higher than aim (0.015% for both). As discussed below, the desphosphorization reaction is very sensitive to the carbon content in the steel, and, as it has been shown, \( \tau_{MC} = 0 \) correlates with higher values of TD1 C.

Finally, it is useful to know that the oxygen flow rate and lance height profiles (not shown here) were steady and very similar in the final minutes of all heats. This observation would reduce the likelihood of the possibility that the measured peak in the particle emissivity was caused by the operator’s changes to the oxygen lance height towards the end of each heat.

5.3.2 Relevant Points around the Meltshop Practice of Successful Heats
The main factors that determine success for a heat in a BOF are, in order of importance, bath temperature, carbon and phosphorus content. Sulphur is also relevant, but it is typically addressed at other stages in the steelmaking process, such as the desulphurization station, since not much can be done at the BOF in terms of its removal [1].

A typical range for the turndown temperature is within ±15°C of the target. After the oxygen blow is finished, the temperature and dissolved oxygen in the steel are measured with an immersed probe and used as inputs to the BOF alloy model. The model is run to determine the additives required to recarburize the heat with coke, deoxidize the steel with silicon and aluminum, and achieve the desired alloy composition with components such as chromium and manganese. The model takes into account the endothermicity or exothermicity of the reactions due to the additions. The fact that TD1 T was higher than aim by as much as +150 °C (Figure 5-3b) and that the average difference for the eight heats was +115 °C (Table 5-1), suggests that two possible causes be inspected: the accuracy of the scales to weigh the scrap, hot metal and fluxes, and the static charge model that sets the amount of O₂ to be blown. If the cause of the temperature rise was the latter, it would lead to an over-injection of oxygen in heats 3, 4 and 8.
Tracking the particle emissivity peak would signal the operator that the end of the blow should be approaching, and this feature would act as a real-time check with which to gauge or adjust the pre-set O$_2$ volume to be blown.

In terms of carbon, it is common practice to turndown at values below aim. Some operators aim for TD1 C = 0.03-0.04% for low carbon heats, and even sometimes for high carbon heats. One can always recarburize the heat at tap, but the phosphorus target is usually linked to low carbon levels. Carbon and phosphorus are related via the dissolved oxygen in the metal and the FeO level in the slag. The higher the carbon content, the lower the dissolved oxygen in the metal and the lower the FeO in the slag. A certain level of FeO (about 20-25%) is required in the slag for the dephosphorization reaction to take place. If the slag is “dry” –meaning that the FeO level is low making the slag less fluid and decreasing its oxidation potential– phosphorus removal will not be as efficient. This poses a problem since the BOF is the only station where dephosphorization can be accomplished. For example if the aim carbon is 0.08% and the turndown is exactly at 0.08%, the dissolved oxygen in the metal will be around 400 ppm. This oxygen level correlates with about 18% FeO in the slag, making it a “dry” slag in terms of dephosphorization. Thus although the carbon target is achieved, the removal of phosphorus will be jeopardized. If instead, and for the same aim carbon of 0.08%, at turndown TD1 C = 0.04%, the dissolved oxygen will be around 700 ppm and the FeO in the slag will be 20%, which will be more suitable for dephosphorization (the removal of phosphorus, apart from the FeO level, depends on the slag basicity and its V-ratio \(\equiv \text{CaO}/\text{SiO}_2\), the initial P level and the oxygen injection configuration). The molten steel would then be recarburized at tap with coke and other additives. With these considerations in mind, it is evident why heats 6 and 7 (both with \(\tau_{MC} = 0\), and TD1 C = 0.078% in heat 6 and TD1 C = 0.170% in heat 7) displayed higher TD1 P values, which were above aim in this case. The other six heats with \(\tau_{MC} > 0\) and lower TD1 C yielded
smaller TD1 P and were able to meet their aim values (Figure 5-3c). The average phosphorus reduction in the refined steel relative to the hot metal input for the two heats with $\tau_{MC} = 0$ was 59.1%, whereas for the other heats with $\tau_{MC} > 0$, the reduction was significantly higher (86.5%).

### 5.3.3 Oxygen End-Point Strategy for Low Carbon Heats

Figures 5-2 and 5-3a suggest that a few seconds after the particle emissivity peaks, that is $\tau_{MC} > 0$ s, the first turndown carbon of the low carbon heats had reached minimum carbon (average = 0.037%). This carbon concentration was sufficient to ensure enough FeO in the slag to oxidize phosphorus to an average TD1 P of 0.006%. The bath temperature target was met in all cases by at least 59°C (heat 5; Table 5-1; average for low carbon heats = 103°C). In the present tests the ratio of hot metal temperature to aim temperature was 0.82 (average for low carbon heats; Table 5-1). Checking if this ratio (HM T/Aim T) falls around 0.82 and then monitoring the bath temperature with the use of a sublance a few minutes before the end of the blowing period would allow the operator to estimate if the temperature target will be met. Assuming that the temperature is on target, the suggested strategy would be for the operator to wait for the particle emissivity to peak, which is also correlated with a drop in $T_p$, and stop the oxygen injection at $\tau_{MC} \approx 30$ s. With the help of the immersion probe at the end of the heat, the operator could run the alloy model to recarburize, deoxidize and alloy the steel. As mentioned above, these are preliminary results and it would be worthwhile to conduct further studies in order to confirm the trends that have been here reported.

It would also be interesting to develop in-house correlations of $\tau_{MC}$ and TD1 T, with which to gauge by how much to extend the blow if necessary. The specific value in $\tau_{MC}$ (here 130 s) after which the temperature rise reaches a plateau with increased oxygen injection could also be determined (Figure 5-3b).
Regarding the high carbon heats, the two heats with $\tau_{MC} > 0$ s indicate that TD1 C is close to minimum carbon and that TD1 P was lower than for the two heats with $\tau_{MC} = 0$ s (heats 1 and 2). The relationship between TD1 T and $\tau_{MC}$ is not clear at this point: even though $\tau_{MC}$ was longer by 29 s for heat 2 relative to heat 1, the lower TD1 T for heat 2 (24 °C) could simply reflect the initial difference in hot metal temperatures, which was lower by 16 °C for heat 2. However more measurements would confirm these findings for high carbon heats.

Finally, the benefits from an optimized end-point strategy for oxygen control in low carbon heats are:

(a) the productivity (ton of steel per day) is increased.

(b) energy usage per ton of steel produced is decreased.

(c) the metal yield is increased.

(d) natural resources per ton of steel are reduced.

(e) direct and indirect CO$_2$ emissions per ton of steel are minimized, especially when fossil fuels are used in the production of high-purity oxygen.

5.4 Conclusions

The successful application of the IR sensor to monitor the off-gas at the mouth of a full-scale converter during low and high carbon heats yielded the following findings:

1. The real-time (every 2 s) particle emissivity profile captures well the transition in decarburization mechanism from oxygen flow rate control to mass transfer control regimes. The feature that announces the transition is a well defined peak in $\varepsilon_p$, which is also correlated with a drop in the particle brightness temperature ($T_p$).
2. A new variable called the *minimum carbon time* ($\tau_{MC}$) was helpful in interpreting the first turndown properties (temperature, carbon and phosphorus) for eight heats. This variable measures in seconds the oxygen injection time from the moment that the particle emissivity reached its peak. If the oxygen lance went off-line before $\varepsilon_p$ peaked, $\tau_{MC}$ is defined as zero.

3. The off-gas evolution from the converter drops sharply either when a heat reaches the mass transfer control regime ($\tau_{MC} > 0$ s) or when the oxygen lance is turned off ($\tau_{MC} = 0$ s). In both cases, the off-gas temperature and the particle brightness temperature decrease significantly because no more radiation from the off-gas and entrained particles reaches the IR sensor.

4. For the six heats with $\tau_{MC} > 0$ s, the turndown carbon and phosphorus levels were both lower than for the two heats with $\tau_{MC} = 0$ s, and below aim. The turndown phosphorus for the two heats with $\tau_{MC} = 0$ s, were above aim.

5. The four low carbon heats, which had $\tau_{MC}$ values greater than zero, displayed steady minimum carbon (average = 0.037%) and phosphorus (average = 0.006%) turndown compositions despite continued oxygen injection. The bath temperature in the four low carbon heats was positively correlated with increasing values of $\tau_{MC}$. The temperature rise, caused by the oxidation of iron in the bath, leveled off after $\tau_{MC}$ reached 130 s.

7. A strategy to obtain low and reliable carbon and phosphorus levels during low carbon heats might be for the operator to monitor the peak in particle emissivity and to turn off the oxygen lance after approximately 30 s. This assumes that provisions are made to meet the temperature target.
8. The pattern that emerges from correlating the first turndown properties with $\tau_{MC}$ appears to be consistent, yet it is based on a field test consisting of only eight heats. Thus these encouraging results are to be taken as preliminary and in need of further verification.

### 5.5 References


6 Conclusions

This chapter summarizes the main contributions of this doctoral project.

The overall contribution was the implementation of procedure to measure remotely gas temperature and spectral particle emissivity in high temperature gas-particle industrial streams. The range of retrievable temperatures was from 700K to 2150K with an estimated error of 2%, depending on the particle size and volume fraction. Particle emissivities were measured between 0.1 and 0.9 with an estimated error of 20%. The successful completion of this project included building a prototype (the IR sensor), developing the retrieval algorithms and testing the prototype at two industrial sites.

First and second generation prototype IR sensors were built and calibrated. The sensor consists of light collecting optics, a light dispersion element (grating spectrometer) and a linear-array pyroelectric detector with 64 pixels. The unit is powered either from AC power or a 9 volt battery. The first prototype was tested at coal-fired boiler (Nanticoke; Chapter 3) and the second one at a steelmaking furnace (U.S. Steel Canada, formerly Stelco; Chapters 4 and 5). The sensor recorded in real-time (every second) the radiance versus wavelength profile in the 3.5 to 5 \( \mu m \) region from which gas temperature and particle emissivity were retrieved.

The temperature was calculated by a least-squared minimization algorithm from two spectral regions: the particle-only region (3.8-4.1 \( \mu m \)) and the gas-particle region (4.56-5.7 \( \mu m \)). From the measured radiance profiles in these regions the initial particle brightness temperature \( T_p \) and the initial physical temperature of the gas-particle stream \( T_{g+p} \) were estimated, respectively.

The particle emissivity \( \varepsilon_p \) in the mid-infrared (at \( \lambda = 3.95 \mu m \)) was calculated from an analytical solution to the Radiative Transfer Equation (RTE) for a semi-infinite column of
suspended particles. The asymptotic solution gives the emerging radiance from the particle cloud and was based on the method of embedded invariance. The final equation related \( \varepsilon_p \) to the effective particle emissivity (\( \varepsilon_{\text{eff}} \))—a secondary variable derived from \( T_p \) and \( T_{g+p} \)—and the cosine of the angle between the line of sight of the sensor and the normal to the gas-particle column (\( \mu \)).

The magnitude of the gas-particle temperature and the particle emissivity could be affected by particle scattering. A new analytical solution to the RTE was developed to include the presence of an absorbing-emitting gas in the particle cloud. The asymptotic solution can be used to describe the emerging radiance in the gas-particle wavelength region (4.56-5.7 \( \mu \)m) for optically thick gas-particle clouds, which implies long path lengths and high concentrations. The method of embedded invariance was again employed. From this formula the conditions that warrant the correction of \( T_{g+p} \) and \( \varepsilon_p \) were established. An iterative procedure to correct gas-particle temperature and particle emissivity was applied to estimate the average error in the datasets from the coal-fired boiler and the steelmaking furnace. The correction for particle scattering effects is best carried out as a post-processing step (since real-time particle size and volume fraction, and gas concentration information is not readily available).

From the analysis of the time series data from the test at a steelmaking furnace (called a Basic Oxygen Furnace or BOF) that spanned eight heats (or batches), an end-point control strategy for oxygen injection was proposed. The strategy was elucidated from the correlations of a new variable introduced here (the minimum carbon time, \( \tau_{\text{MC}} \)) with the steel bath parameters at the end of each heat. The minimum carbon time measures the time between a noticeable peak in the particle emissivity and the time when the oxygen injection stopped. It was observed that for low-carbon heats, the carbon and phosphorus concentrations of the steel bath remained steady for \( \tau_{\text{MC}} > 0 \) s, whereas the steel bath temperature increased due to the oxidation of iron. The explanation is consistent but requires further verification in a BOF facility.
6.1 Future Work

The next step in the commercial deployment of the IR sensor—whose intellectual property has been licensed by the author and his supervisor to Tenova Goodfellow Inc. through a commercialization agreement—would be to harden it to be used continuously at a BOF facility. Some suggestions to avoid the drift in wavelength calibration appear in Appendix 2. Since the oxygen injection end-point strategy shows promise, extended testing with the IR sensor at the BOF facility is recommended.

It would be interesting to relate the change in the particle emissivity towards the end of the heat (Figure 5-2) to the composition of the particles. One hypothesis is that between the oxidation of carbon and iron at the end of decarburization, another species in the melt reacts preferentially with oxygen for a brief period of time (a few tens of seconds). To validate this hypothesis, particle samples should be taken before, during and after the emissivity peak. Comparing their composition may explain the observed spike in particle emissivity.

New tests could be carried out at a BOF to measure the exhaust gas temperature and the particle emissivity with the IR sensor at the mouth of the converter, and the exhaust gas composition (CO, CO₂, H₂ and O₂) with Tenova Goodfellow Inc.’s EFSOPTM analyzer positioned further downstream along the exhaust duct. Superimposing the CO₂, CO, temperature and particle emissivity traces may help to identify an even more robust marker for carbon depletion in the bath than \( \tau_{MC} > 0 \) s.

A new version of RADCAL [2]—a one-dimensional RTE solver used extensively in this project—could be developed by porting the current FORTRAN code to Matlab or Java and by adding usability through a proper user interface. The spectroscopic data for some combustion gases could be updated with help from the Laboratoire d’Énergétique Moléculaire et Macroscopique, Combustion (EM2C) in France. The new RADCAL version could include the calculation of the
emerging radiance from large gas-particle columns over all wavelengths provided that the particle loading is high enough to satisfy the optically-thick condition. Thus, even though the spectral region of the wings of the gas bands may be optically thin despite the long path lengths and high gas concentrations, Equation 3-26 would still be valid because the particle component makes the gas-particle cloud an optically thick system.
References

These references apply to the Introduction and Conclusions chapters and the Appendices. Chapters 2-5 have their own references at the end of each chapter.


Appendix 1 The Difficulty of Measuring CO and CO\textsubscript{2} When Particles are Present in Industrial, High-Temperature Gas-Particle Streams with Low-Resolution Emission Spectroscopy in the Mid-Infrared (4-5 \textmu m)

This appendix comments on the difficulty of relating the emission profile of CO and CO\textsubscript{2} in high-temperature gas-particle streams to variations in the gas concentration. The emission profile is measured by a low-resolution sensor (64 pixel linear array with pixel width of 20 nm) between 4 and 5 \textmu m. The more complicated problem of varying concentration and temperature concurrently is not addressed here given the present findings.

The main difficulty is the lack of contrast between the gas radiance emission and the broadband particle emission. This lack of contrast is caused by:

- the saturated nature of the CO and CO\textsubscript{2} emission bands, with the CO\textsubscript{2} band saturating more easily (i.e., at shorter path lengths and smaller concentrations). Gas saturation is, on the other hand, the property that allowed us to measure the temperature of the gas-particle stream (Chapter 2). The only viable region becomes the right wing of the gas emission band (longer wavelengths, $\lambda \approx 4.8$ \textmu m).

- the broadband emission of the particles entrained by the gas, even for particles with modest emissivities ($\varepsilon_p \approx 0.4$). The effect of this background is very noticeable as shown in Figure A1-1 for CO (CO\textsubscript{2} behaves similarly). Thus particle broadband emission blurs the already saturated gaseous profile making it difficult to distinguish concentration changes.
Figure A1-1. RADCAL [2] emission profiles of 10% and 50% CO in air (1 atm, 1600K) with and without (“no BB” lines) broadband particle emission ($\varepsilon_p = 0.4$) modeled by a background temperature of $T_p = 1535K$.

-the likely lowering of the overall radiance due to particle scattering effects (as was the case at the coal-fired boiler of Chapter 2 and the steelmaking furnace of Chapter 4).

-the presence of both CO and CO$_2$ with concentrations changing in time and whose emission profiles overlap between 4.5 and 5 μm, decreases even further the signal contrast (Figure A1-2). The possibility of differentiating the three lines in Figure A1-2 is very small for a device like the IR sensor.
Figure A1-2. RADCAL [2] emission profiles at 1 atm and 1600K of 20% CO₂ in air, and to mixtures of CO and CO₂ (balance = air) with broadband particle emission ($\varepsilon_p = 0.4$) modeled by a background temperature of $T_p = 1535K$. 
### Appendix 2 Materials and Operation of the IR Sensor and Suggestions for the Hardening of the Prototype

Table A2-1 lists all materials used in the building and calibration of the current version of the IR sensor prototype.

<table>
<thead>
<tr>
<th>Module name</th>
<th>Components</th>
<th>Supplier</th>
</tr>
</thead>
<tbody>
<tr>
<td>Collecting Optics</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Focusing Lens</td>
<td>CaF₂ plano-convex lens; Diameter = 25.4 mm; Focal Length = 75 mm</td>
<td>Thorlabs</td>
</tr>
<tr>
<td>Long-Pass Filter</td>
<td>Transmission: 3.60–7.13 µm</td>
<td>Spectrogon</td>
</tr>
<tr>
<td>Lens Tube</td>
<td>Diameter = 25.4 mm; Length = 50 mm</td>
<td>Thorlabs</td>
</tr>
<tr>
<td>Chopper Housing</td>
<td>Also serves to hold lens tube</td>
<td>MIE Machine Shop</td>
</tr>
<tr>
<td>Dispersion Optics</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oriel MS125 Spectrometer</td>
<td>Focal length = 120 mm</td>
<td>Newport</td>
</tr>
<tr>
<td>Diffraction Grating</td>
<td>75 lines/mm</td>
<td>Newport</td>
</tr>
<tr>
<td>Entrance Slit</td>
<td>Height = 3 mm; Width = 200 µm</td>
<td>Newport</td>
</tr>
<tr>
<td>Pyroelectric Detector</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IR Microsystems μray64 Kit*</td>
<td>64 Pixel Detector; Chopper; Chopper Driver</td>
<td>Electro Optical Components</td>
</tr>
<tr>
<td>Detector-Spectrometer Flange</td>
<td>MIE Machine Shop</td>
<td></td>
</tr>
<tr>
<td>9V Power Supply</td>
<td>Connects to a 120V AC outlet</td>
<td>Hardware store</td>
</tr>
<tr>
<td>Data Acquisition and Analysis</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hardware: Inspiron 600m Laptop</td>
<td>Dell</td>
<td></td>
</tr>
<tr>
<td>Software: Windows XP; Matlab; Labview</td>
<td>MIE Computer Store</td>
<td></td>
</tr>
<tr>
<td>NEMA Box</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sapphire Window</td>
<td>Diameter = 75 mm; Thickness = 2 mm</td>
<td>Edmund Optics</td>
</tr>
<tr>
<td>Base Flange; Window Flange; Several Mechanical Anchors</td>
<td>MIE Machine Shop</td>
<td></td>
</tr>
<tr>
<td>Wavelength Calibration Kit</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Narrowband Filters (2)</td>
<td>Centre wavelengths = 3.9 µm and 4.6 µm</td>
<td>Spectrogon</td>
</tr>
<tr>
<td>Filter Holders (2) and Rings (4)</td>
<td></td>
<td>Thorlabs</td>
</tr>
<tr>
<td>IR Source</td>
<td>Uniline Ignitor Kit</td>
<td>Reliable Parts Ltd.</td>
</tr>
<tr>
<td>Power Supply for IR Source**</td>
<td></td>
<td>McMaster-Carr</td>
</tr>
<tr>
<td>Stage for IR Source</td>
<td></td>
<td>Edmund Optics</td>
</tr>
<tr>
<td>Radiance Calibration Kit</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mikron M330 Blackbody Calibration Source**</td>
<td></td>
<td>Spectral Applied Research</td>
</tr>
</tbody>
</table>
* IR Microsystems announced in 2007 that they will discontinue this kit until more demand is found; ** Not property of the University of Toronto Combustion Group.

Table A2-1 Bill of materials and suppliers for the building and calibration of the current version of the IR sensor prototype.

Data Acquisition and Analysis

Raw data in terms of counts versus pixel per second is the basic output from the IR Microsystems µarray64 Kit. It is recorded in real time by a LabView program developed at the University of Toronto, called IRRead. This program displays the raw data and performs its analysis in real time. This is done by interfacing IRRead with custom-made Matlab functions that perform the retrieval algorithms. IRRead requires as inputs a wavelength calibration file and a radiance calibration file. Both calibration files are in Matlab format. There is a custom-made Wavelength Calibration program in LabView that directly outputs the Matlab file needed by IRRead. The same is not true for Radiance Calibration, which requires further work in order to be streamlined and documented. The output file of IRRead is a text file that saves the raw and analyzed data in real time. The analysis takes approximately one second, so that each data vector (raw data + analyzed data) is added to the output file every 2.024570344 seconds. If the IR sensor will be running constantly in a BOF facility, code should be written to plot only the relevant heat data. Currently this step is done manually in Matlab. We have only experience with IRRead output files that span over two and a half days. The size of these text files are under 100 MB.

Prototype Hardening

Dust Accumulation and Alignment

The number of counts can be diminished either by dust build-up on the sapphire window of the NEMA box or by misalignment (i.e., the field of view of the IR sensor is not completely filled by the gases of the combustion gap). Less number of counts translates into lower retrieved temperatures. The current design allows for instrument air to come into the NEMA box and blow by the sapphire window to keep it clean. Five psig is the suggested pressure. We also tried 20
psig (after running at 5 psig for about one day at the BOF facility) but this seemed to affect negatively our results. Maintenance schedules for the cleaning of the sapphire window and the metal tube on the furnace skirt should be developed and tested.

_Vibration and Wavelength Calibration_

Mechanical vibrations will shift slightly the relative position between the pyroelectric detector and the spectrometer, even though the flange between the two has been sufficiently reinforced already. This shift is known to cause a change of up to 80 nm in the wavelength range covered by the 64 pixels, keeping all other factors constant. Since a change of 80 nm will affect \( T_{g+p} \) more than \( T_p \) (the particle-only region is wider than the gas-particle one), it is worth investigating to replace the spectrometer with a fixed linear variable filter glued on the window of the pyroelectric detector (a good initial contact would be IR Microsystems).

_Chopper Failure and Dead Pixel_

The motor that drives the chopper has been known to stop randomly for less than 10 seconds. One of the 64 pixels is malfunctioning. Its behaviour is currently masked by linearly interpolating the values of the neighbouring pixels.
Appendix 3 Nature of the Collaboration with other Members of the Combustion Group

This appendix explains the contribution of each of the members of the Combustion Group on the IR sensor project. The wavelength and radiance calibration and the basic temperature retrieval algorithm were primarily led by M.A.Sc. graduate Rebecca Saari [3], who also provided valuable assistance (error analysis calculations, proof-reading) in the drafting of the second chapter. On the Nanticoke test I worked with Sameh El-Batraoukh (former Ontario Power Generation employee and M.Eng. student) responsible for the logistics of the trip and Reza Mani (former post-doctoral fellow at the Combustion Group) who turned the IR sensor into a stand-alone unit through a series of batteries. In the BOF test Reza Mani built the heat-resistant enclosure for the IR sensor (NEMA box), and Rebecca Saari and Fut Yang (undergraduate student who worked at the Combustion Group in summer 2006) incorporated the retrieval algorithms into the data acquisition package so that the initial gas temperature and particle emissivity could be seen in real-time. The author and his supervisor are directly responsible for the conceptual analysis of the sources of radiation in the 3.5-5 µm region; the retrieval of particle emissivity from the particle brightness temperature and the physical temperature of the gas-particle medium; the theoretical developments behind particle scattering effects on the magnitude of $T_{g+p}$ and $\varepsilon_p$; and for the process control strategy for oxygen injection in a BOF.