# Radiative Heat Transfer in Oxy–Fuel Steam Generators

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# Abstract

The purpose of this dissertation is to assess the impact of oxy-fuel combustion on radiative heat transfer in steam generators and its calculation. Radiative transfer in a lignite-fired 30 MW oxy-fuel furnace is modeled on the basis of measured gas temperature and concentration of major radiating gas species by Monte Carlo ray tracing, with the intent of model calibration. Simulation results are evaluated and compared to measured values of incident radiative heat flux. This work contributes to the evaluation of radiative transfer in large furnaces with respect to optical path length. Adaptation of a global emissivity model is presented and compared to other emissivity correlations fitted to high concentration of radiating gas species. Recommendations on coupling global emissivity and radiation model with spectrally averaged absorption coefficients are given. Finally, the adapted emissivity model is applied to CFD simulation of the regarded furnace in order to estimate its influence on gas temperature and radiative heat flux.

This dissertation should be of interest to process and combustion engineers, in the fields of fossil fuel fired power generation and carbon dioxide capture technology. It should also be of interest to software engineers, and practitioners of combustion and in particular radiative transfer simulation.

# Kurzfassung

In Zusammenhang mit der Vermeidung von Kohlendioxidemissionen sind fossil befeuerte Kraftwerke in den Fokus für die Anwendung von Technologien des carbon capture and storage (CCS) gerückt. Diese sind dazu geeignet, das bei der Verbrennung von fossilen Brennstoffen unweigerlich entstehende Kohlendioxid abzuscheiden. Um eine Abgabe des Kohlendioxids an die Atmosphäre zu verhindern, soll dieses beispielsweise in bestimmten geologischen Formationen gespeichert werden. Hierfür muss aus Sicht der Kraftwerkstechnik ein möglichst hoch konzentrierter Kohlendioxidstrom erzeugt werden, damit der Aufwand für die zur Speicherung notwendige Aufbereitung und Kompression möglichst gering bleibt. Das Abgas fossil befeuerter Kraftwerke enthält aber lediglich einen von der Art des Brennstoffs abhängigen Anteil von Kohlendioxid. Bei Kohlekraftwerken besteht das Rauchgas bei der Verbrennung mit Luft üblicherweise zu 10 bis 15 Vol.-% aus Kohlendioxid. Zur Bereitstellung eines hochkonzentrierten Kohlendioxidstroms sind im Wesentlichen drei Technologien geeignet: eine nachgeschaltete Rauchgaswäsche (post-combustion), eine vorgeschaltete Brennstoffvergasung mit CO<sub>2</sub>-Abscheidung aus dem Produktgas (pre-combustion) und die Verbrennung mit Sauerstoff anstelle von Luft (oxy-fuel).

Diese Arbeit bezieht sich auf das letztgenannte Verfahren, bei dem der Brennstoff anstelle von Luft mit technisch reinem Sauerstoff verbrannt wird. Der Sauerstoff muss hierfür zunächst in einer Luftzerlegungsanlage hergestellt werden. Bei der Verbrennung mit dem aus der Luft gewonnenen Sauerstoff wird eine Verdünnung des Rauchgases mit dem Stickstoff der Luft verhindert. Es entsteht dann ein Rauchgas, das überwiegend aus Kohlendioxid und Wasserdampf besteht, wobei letzterer durch Kondensation weitgehend aus dem Rauchgas entfernt werden kann. Es entsteht auf diese Weise ein Rauchgasstrom mit sehr hoher Konzentration von Kohlendioxid und geringen Anteilen von verunreinigenden Gasen wie Sauerstoff, Stickstoff, Argon, Wasserdampf und Stickoxiden. Da sich unter diesen Umständen auch der Volumenstrom des Rauchgases stark verringert, ist es notwendig, einen Teil des Rauchgases in die Brennkammer zurückzuführen, da ansonsten die aus der Verbrennung entstehende Wärme nur auf eine sehr geringe Menge Rauchgas übergehen und zu sehr hohen Rauchgastemperaturen in der Brennkammer führen würde. Aus diesem Grund wird ein Teil des Rauchgases zur Kühlung in die Brennkammer geleitet. Mittels der Menge rezirkulierten Rauchgases kann die Rauchgastemperatur beeinflusst werden.

Die Wärmeübertragung in Oxy-fuel-Dampferzeugern ist Gegenstand der Forschung, da die veränderte Rauchgasatmosphäre andere thermodynamische Eigenschaften aufweist. Diese ergeben sich aus den unterschiedlichen physikalischen Eigenschaften von Kohlendioxid und Wasserdampf gegenüber Stickstoff, aus welchem das Rauchgas im Fall der konventionellen Verbrennung mit Luft hauptsächlich besteht. Im Gegensatz zu Stickstoff sind Kohlendioxid und Wasserdampf als heteroatomige Gase in der Lage elektromagnetische Strahlung im Bereich der Wärmestrahlung zu absorbieren und zu emittieren. Dadurch erhöht sich die Absorptivität bzw. Emissivität des Rauchgases bei der Oxy-fuel-Verbrennung im Vergleich zur Verbrennung mit Luft, da sie von der Konzentration emittierender bzw. absorbierender Gase abhängig ist. Hierbei muss aber beachtet werden, dass Strahlungsprozesse in Dampferzeugern von Kohlekraftwerken häufig von den kontinuierlich über das Spektrum Wärmestrahlung emittierenden und absorbierenden Ruß-, Asche-, und Kohlepartikel dominiert werden.

In dieser Arbeit wird ein Dampferzeuger mit 30 MW thermischer Leistung untersucht, der nach dem Oxy-fuel-Verfahren betrieben werden kann. Die Anlage verfügt über alle für den Prozess wesentlichen Komponenten und ist eine der weltweit größten Anlagen dieser Art. Dies ist im Hinblick auf die im Vergleich mit Labor- und Versuchsanlagen große mittlere Weglänge in der Brennkammer des Dampferzeugers von Bedeutung. Es werden Messungen der Rauchgastemperatur (Abschnitt 3.3.1) und der Gaszusammensetzung (Abschnitt 3.3.2) in der Brennkammer durchgeführt, um die Verhältnisse im Feuerraum in einer Simulation richtig abbilden zu können (Abschnitte 4.2.1 und 4.2.2). An der Vorderwand des Dampferzeugers wird die auf die Feuerraumwand auftreffende Wärmestrahlung gemessen (Abschnitt 3.2.4). Diese Messungen werden für den Vergleich mit den aus der Simulation erzielten Ergebnissen zur Wärmeübertragung verwendet (Kapitel 5).

Die Berechnung der Wärmestrahlung erfordert die näherungsweise Lösung einer Transportgleichung, die den Strahlungsvorgang entlang eines Intensitätsstrahls beschreibt (siehe Abschnitt 2.1). Diese Gleichung wird auch als *Strahlungstransportgleichung* bezeichnet. Die näherungsweise Berechnung der Wärmeübertragung durch Strahlung wird mit Hilfe einer Strahlverfolgung durchgeführt. Die Strahlverfolgung beruht auf der *Monte-Carlo-Methode*, bei der die Wärmestrahlung als eine hohe Anzahl von einzelnen Strahlen mit einem inkrementellen Energieinhalt angesehen wird. Ausgangsposition, Richtung, Absorption, Reflexion und Streuung eines Strahls werden dabei anhand von Zufallszahlen, die eine gleichmäßige Häufigkeitsverteilung aufweisen, bestimmt.

Der Rechenaufwand für die näherungsweise Lösung der Strahlungstransportgleichung ist aufgrund der Komplexität der Strahlungsvorgänge extrem hoch, so dass für die Anwendung in der Verbrennungstechnik die graue Formulierung der Strahlungstransportgleichung (Abschnitt 2.1.1) verwendet wird. Dabei ist es üblich, die Abhängigkeit der Wärmestrahlung von der Wellenlänge zu vernachlässigen (spektrale Mittelung), Absorptions- und Emissionskoeffizient des Gases als gleich und lokales thermodynamisches Gleichgewicht anzunehmen. Für Letzteres muss der betrachtete Feuerraum in eine Anzahl von Volumenelementen unterteilt werden, in denen alle intensiven thermodynamischen Größen als konstant angenommen werden können. Außerdem gilt die graue Formulierung der Strahlungstransportgleichung nur, wenn im Feuerraum keine oder lediglich geringe Gradienten dieser Größen vorhanden sind.

Es gibt zahlreiche Modelle zur Berechnung einer intensiven Größe, die die Fähigkeit Wärmestrahlung zu absorbieren bzw. zu emittieren beschreibt. Diese Größe wird Emissivität genannt. Die vorhandenen Modelle sind in der Regel für die Berechnung der Rauchgasemissivität von Gasgemischen mit deutlich geringeren Konzentrationen von Kohlendioxid und Wasserdampf als bei der Oxy-fuel-Verbrennung geeignet. Ein häufig gebrauchtes Modell ist das *weighted-sum-of-gray-gases-model* (WSGGM), bei dem die Emissivität als gewichtete Summe der Emissivitäten mehrerer grauer Gase und meistens eines für die Wärmestrahlung transparenten Gases berechnet wird. Die einzelnen Summanden stellen in diesem Modell über bestimmte spektrale Abschnitte gemittelte Strahlungseigenschaften dar und die Gewichtung ihren spektralen Anteil an der Wärmestrahlung. In dieser Arbeit wird zur Simulation der Strahlungswärmeübertragung ein WSGGM mit Gewichtungsfaktoren und Absorptionskoeffizienten eingesetzt, bei welchem beide Größen Funktionen des Verhältnisses der Molanteile von Wasserdampf zu Kohlendioxid sind.

Darüber hinaus wird zur Lösung der Strahlungstransportgleichung eine extensive Zustandsgröße, der spektral gemittelte Absorptionskoeffizient, benötigt, der im Sinne der grauen Formulierung der Strahlungstransportgleichung auch der Emissionskoeffizient ist. Für die Berechnung der Wärmeübertragung in Feuerräumen sind weder der mittlere Absorptionskoeffizient nach Planck für optisch dünne Medien noch der mittlere Absorptionskoeffizient nach Rosseland für optisch dichte Medien geeignet. Üblicherweise wird für die Simulation von Verbrennungsvorgängen ein *effektiver*  Absorptionskoeffizient berechnet, der eine Ad-hoc-Berechnung aus dem Lambert-Beerschen Gesetz darstellt. Der mittlere Absorptionskoeffizient nach Patch bietet hingegen eine rigorose Lösung dieses Problems, bei der die Selbstabsorption von im Gas emittierter Strahlung durch das Gas Berücksichtigung findet. Der effektive Absorptionskoeffizient und der Absorptionskoeffizient nach Patch unterscheiden sich bei der Verbrennung mit Luft und den dabei auftretenden Molanteilen von Wasserdampf und Kohlendioxid nur geringfügig voneinander. Aufgrund höherer Molanteile von Kohlendioxid und Wasserdampf kann für die Oxy-fuel-Verbrennung gezeigt werden, dass der effektive Absorptionskoeffizient deutlich vom Absorptionskoeffizienten nach Patch abweicht.

Neben den Strahlungseigenschaften muss für jedes Volumenelement auch die Temperatur bestimmt werden. Dies geschieht üblicherweise durch eine iterative Lösung der Strahlungs- und Energietransportgleichung. Dies ist jedoch in diesem Fall nicht notwendig, da die Temperaturverteilung durch Annäherung von Messwerten mittels geeigneter analytischer Näherungsfunktionen bestimmt wird. Auf diese Weise werden Einflüsse anderer chemisch-physikalischer Modelle, die für die Simulation einer Verbrennung notwendig sind, eliminiert und die Berechnung bleibt auf die Lösung des Wärmestrahlungsproblems begrenzt.

Der Absorptionskoeffizient des Gases bei der Oxy-fuel-Verbrennung ist aufgrund der höheren Kohlendioxid- und Wasserdampfkonzentration im Rauchgas höher. In Bereichen mit hoher Konzentration von Ruß-, Kohle- oder Aschepartikeln spielt der Gasabsorptionskoeffizient aber nur eine untergeordnete Rolle. Der berechnete Gasabsorptionskoeffizient ist bei hoher Konzentration von Feststoffpartikeln deutlich niedriger als der berechnete Partikelabsorptionskoeffizient. Er kann jedoch in der Nähe der Feuerraumwand, wo die Partikelkonzentration niedrig ist, bei der Oxyfuel-Verbrennung auch bis zu 1,5-fach höher sein als der Partikelabsorptionskoeffizienten grundsätzlich empfehlenswert.

Nimmt man für eine Einschätzung der Auswirkung der höheren Konzentration von Kohlendioxid und Wasserdampf an, dass die Temperatur– und Partikelverteilung in der Brennkammer bei der Verbrennung mit Luft und bei der Oxy– fuel–Verbrennung genau gleich sei, so führt die erhöhte Konzentration der beiden Gase zu einer Erhöhung der Gasemissivität um 30 % und zu einer Erhöhung der Wandwärmeströme um durchschnittlich 7,3 % gegenüber der Verbrennung mit Luft. Die Simulation zeigt signifikante Unterschiede bei den Wandwärmestromdichten, je nachdem ob der effektive Absorptionskoeffizient oder der auf einer rigorosen Herleitung beruhende Absorptionskoeffizient nach Patch in der Strahlungstransportgleichung eingesetzt wird. Der effektive Absorptionskoeffizient ist, insbesondere bei der Verwendung eines Emissivitätsmodells für die Verbrennung mit Luft, deutlich höher als der Absorptionskoeffizient nach Patch und führt zur Berechnung von zu hohen Wandwärmestromdichten im Vergleich zur Messung. Die auf diese Weise auftretende Überschätzung des Absorptionskoeffizienten des Gases war bislang bei der Verbrennung mit Luft von untergeordneter Bedeutung, weil bei dort üblichen Partialdrücken von Kohlendioxid und Wasserdampf die Unterschiede zwischen Patch– und effektivem Absorptionskoeffizienten nur gering sind. Die näherungsweise Übereinstimmung der beiden Koeffizienten gilt bei üblichen Weglängen und höheren Partialdrücken von Kohlendioxid und Wasserdampf nicht mehr. Es ist daher nicht empfehlenswert, den effektiven Gasabsorptionskoeffizienten bei der Berechnung der Wärmeübertragung durch Strahlung in Oxy–fuel–Dampferzeugern zu verwenden.

Da bei der Simulation des Strahlungswärmeaustausches in der untersuchten Brennkammer die Verteilung der Gastemperaturen mit Hilfe von analytischen Funktionen angenähert wird, erfordert die Lösung der Strahlungstransportgleichung keine iterative Vorgehensweise, bei der sich aus der Energiebilanz eines jeden Volumenelements eine Temperatur ergibt. Zur Bewertung der Auswirkung der Gasstrahlungseigenschaften bei unbekannter Temperaturverteilung und iterativer Lösung der Strahlungstransportgleichung wird derselbe Feuerraum auch mit der *Computational Fluid Dynamics* (CFD) Software FLUENT 13.0 simuliert. Der Simulation, zunächst ohne Berücksichtigung der Partikelstrahlung, wird zum Einen der effektive Absorptionskoeffizient und zum Anderen der Patch–Absorptionskoeffizient für die Gasstrahlung zu Grunde gelegt. Dabei zeigt sich, dass die auf dem höheren, effektiven Absorptionskoeffizienten beruhende Simulation niedrigere Gastemperaturen und höhere Wandwärmeströme aufweist als die Berechnung auf Basis des Patch– Absorptionskoeffizienten.

Wird jedoch die Absorption und Emission von im Rauchgas vorhandenen Partikeln berücksichtigt, so erhöht sich der Gesamtabsorptionskoeffizient des Rauchgases und es werden höhere Wandwärmeströme sowie niedrigere Gastemperaturen berechnet. Der Gasabsorptionskoeffizient hat nun einen deutlich geringeren Einfluss auf die Berechnungsergebnisse. Der Absorptionskoeffizient der Partikel ist gemäß der Berechnung in FLUENT in weiten Bereichen des betrachteten Feuerraums mindestens doppelt so hoch wie der Gasabsorptionskoeffizient, kann jedoch auch mehr als fünfzehnfach höhere Werte erreichen. Lediglich in Regionen mit sehr geringer Staubkonzentration ist der Gasabsorptionskoeffizient höher als der Partikelabsorptionskoeffizient. Die Wandwärmeströme und Gastemperaturen aus der iterativen Lösung der Strahlungs- und Energietransportgleichung unterscheiden sich nun kaum mehr zwischen der auf dem effektiven und dem Patch–Absorptionskoeffizienten beruhenden Berechnung.

Grundsätzlich machen die CFD–Simulationen deutlich, dass bei einer iterativen Lösung der Strahlungs- und Energietransportgleichung der Absorptionskoeffizient des Gases nicht nur, wie bei der nicht-iterativen Monte–Carlo–Simulation, einen Einfluss auf die Wandwärmestromdichten, sondern auch auf die Gastemperatur hat. Eine Überschätzung des Gasabsorptionskoeffizienten führt daher nicht nur zu einer Erhöhung der berechneten Wandwärmeströme, sondern bewirkt auch eine Absenkung der Temperatur. Da die Temperatur mit der vierten Potenz in den Emissionsterm der Strahlungstransportgleichung eingeht und der Absorptionskoeffizient nur linear, sind durch eine unzureichend genaue Berechnung des Gasabsorptionskoeffizienten und der Gasemissivität nur geringe Fehler der berechneten Temperaturergebnisse zu erwarten. Insgesamt ist es aus diesem Grund möglich, den Strahlungstransport in Feuerräumen mit der durch den Oxy–fuel–Prozess bedingten Konzentrationserhöhung der Gase Kohlendioxid und Wasserdampf in der Rauchgasatmosphäre anhand vereinfachender Modelle mit der selben Qualität abzubilden, wie es für konventionell betriebene Dampferzeuger bislang der Fall war.

# Nomenclature

## Latin Symbols

A	surface area [m <sup>2</sup> ]
	also: total absorptance $[cm^{-1}]$
$A_{int}$	specific internal surface of char $[m^2/kg]$
a	weighting factor [-]
$B_1$	pre–exponential factor in Arrhenius equation [-]
$B_2$	pre–exponential factor in Arrhenius equation [-]
$b_1$	constant $[m^2/kg]$
$b_T$	constant $[K^{-1}]$
c	speed of light $[ms^{-1}]$
$C_0$	constant depending on refractive index [-]
$C_1$	First Planck radiation constant, $3.7417715 \cdot 10^{-16}$ [Wm <sup>2</sup> ]
$C_2$	Second Planck radiation constant, $1.4387770 \cdot 10^{-2}$ [mK]
D	diameter [m]
$D_0$	diffusion rate $[m^2/s]$
d	edge length of volume element [mm]
$d_c$	mesh size of analysis sieve [m]
$\overline{d_c}$	mean particle diameter [m]
$d_{c,m}$	mean linear coal particle diameter [m]
$d_{c,s}$	Sauter diameter [m]
$d_{fa}$	fly ash particle diameter [m]
$d_{ra}$	residual ash particle diameter [m]
E	emissive power $[Wm^{-2}]$
$E_1$	activation energy [J/mol]
$E_2$	activation energy [J/mol]
F	fractional black body function [-]
f	error function [-]
$f_A$	total projected area [m <sup>2</sup> ]
$f_d$	diffuse fraction of radiation [-]

$f_h$	Heywood factor [-]
$f_v$	volume fraction [-]
h	Planck constant $[Ws^2]$
Ι	radiative intensity $[Wm^{-2}sr^{-1}]$
i	number of element in x-direction [-]
j	number of element in y-direction [-]
K	constant [-], number of spectral blocks [-]
k	number of element in z–direction [-]
	also: reaction rate of first order reaction $[1/s]$
$k_B$	Boltzmann constant, 1.380648 $\cdot 10^{-23}$ [WsK <sup>-1</sup> ]
$k_{int}$	intrinsic reaction rate $[kg/m^2s]$
L	optical path length [m]
l	number [-]
$lg_{10}$	decadic logarithm
ln	natural logarithm
$L_p$	Planck mean free path [m]
M	set of numbers
m	mass [kg]
N	number of beams [-]
P	cumulative probability [-]
	also: sum of partial pressures [hPa]
$P_e$	point of emission [-]
$P_{eq}$	equivalent broadening pressure [-]
$P_t$	total pressure [hPa]
p	partial pressure [hPa]
	also: probability [-]
Q	energy [W]
q	heat flux $[kWm^{-2}]$
R	random number [-]
	also: gas constant, $8.3144621~[\mathrm{J/molK}]$
$R_1$	low temperature devolatilization rate $[1/\mathrm{s}]$
$R_2$	high temperature devolatilization rate $[1/s]$
$R_{int}$	intrinsic reaction rate $[kg/m^2s]$
r	spherical coordinate [m]
$r_l$	way of a bundle in a volume element [m]
S	path length [m]
T	temperature [K] or $[^{\circ}C]$

t	time [s]
U	voltage [mV]
u	number of gray gas [-]
V	volume [m <sup>3</sup> ]
X	dummy variable [-]
$x_C$	refractive index [-]
y	volume fraction $[m^3/m^3]$
$y_C$	absorptive index [-]
$z_C$	complex index of refraction [-]
$z_d$	vertical distance from the furnace top [mm]

## Greek Symbols

α	absorptivity [-]
$\alpha_1$	weighting factor [-]
$\alpha_2$	weighting factor [-]
$\beta$	extinction coefficient $[m^{-1}]$
ε	emissivity [-]
$\eta$	line width to spacing parameter [-]
$\eta_{int}$	effectiveness factor [-]
$\Phi$	circumferential angle [rad]
$\Phi_c$	size parameter $[\mathrm{Km}^{-2}]$
$\phi$	azimuthal angle [rad]
$\Psi$	mole fraction ratio of H <sub>2</sub> O/CO <sub>2</sub> , $\psi_{H_2O}/\psi_{CO_2}$ [-]
$\psi$	mole fraction [-]
$\kappa$	absorption coefficient $[m^{-1}]$
$\kappa_e$	effective absorption coefficient [m <sup>-1</sup> ]
$\kappa_p$	Planck mean absorption coefficient $[m^{-1}]$
$\kappa_{pa}$	Patch mean absorption coefficient $[m^{-1}]$
$\kappa_r$	Rosseland mean absorption coefficient $[m^{-1}]$
$\lambda$	wavelength [m]
ρ	reflectivity [-]
	also: density, mass concentration $[kg/m^3]$
σ	scattering coefficient $[m^{-1}]$
$\sigma_c$	Stefan-Boltzmann constant, 5.670373 $\cdot 10^{-8}~[\mathrm{Wm}^{-2}\mathrm{K}^{-4}]$
ν	wavenumber $[m^{-1}]$
	also: polynomial coefficient [-]

$\mu$	polynomial coefficient [-]
Θ	cone angle [rad]
$\theta$	polar angle [rad]
au	transmissivity [-]
$ au_H$	optical depth at band head [-]
Ω	solid angle [sr]
ω	bandwidth parameter $[cm^{-1}]$
X	boundary value of black body fractional function [-]
ζ	partial pressure ratio [-]

## Mathematical Operators

$\Delta$	difference
d	differential operator in Leibniz's notation
$\partial$	partial differential operator
i	imaginary unit
Π	product
$\int$	integral
$\sum$	sum

## Sub - and Superscripts

3 + 1	WSGG3+1
4 + 1	WSGG4+1
abs	absorption
ash	ash
b	black body
C	band center
C	complex
c	coal
$CO_2$	carbon dioxide
conv	convective
corr	corrected
d	distance
e	effective
	also: element
em	emission

EWBM	exponential wide band model
fa	fly ash
g	gas
g + s	gas and soot
gr	gray
$H_2O$	water vapor
i	gas number
int	intrinsic
	also: internal
j	band number
L	lower band limit
max	maximum
min	minimum
n	normal
$O_2$	oxygen
p	Planck
pa	Patch
part	particles
r	residual
r	Rosseland
ra	residual ash
rad	radiative
s	soot
	also: surface element, start
t	total
U	upper band limit
u	gray gas number
v	volume element
vol	volatiles
w	wall
WSGG	weighted sum of gray gases
$\epsilon$	emissivity
ν	function of wavenumber
$\chi$	boundary value of black body fractional function

### Abbreviations

ADF Absorption distribution function m	nodel
--	-------

ADFFG	Absorption distribution function with fictitious gases
CCS	Carbon dioxide capture and storage
CFD	Computational fluid dynamics
CK	Correlated–k model
CKFG	Correlated–k model with fictitious gases
EWBM	Exponential wide band model
LBL	Line–by–line model
LTE	Local thermodynamic equilibrium
NBM	Narrow band model
OFA	Over fire air
RTE	Radiative transfer equation
SLW	Spectral line–based weighted sum of gray gases model
UDF	User–defined function
WBCK	Wide band correlated–k model
WBM	Wide band model
WSGGM	Weighted sum of gray gases model

# 1 Introduction

Production of energy from fossil fuels results in emission of carbon dioxide which is the most important anthropogenic greenhouse gas. Carbon dioxide concentration in the atmosphere has increased from 280 ppm in pre-industrial times to about 400 ppm in 2013. The increase of atmospheric carbon dioxide concentration is mainly attributed to the use of fossil fuels [55]. Energy production from fossil fuels contributes 85 % to the total amount of annual anthropogenic carbon dioxide emissions [119].

Fossil fuels provided 80 % of the worldwide primary energy demand in 2004 with coal being the second largest contribution [119]. In order to decrease anthropogenic greenhouse gas emissions, either energy sources with low or zero carbon dioxide emissions have to be used, e.g. wind or solar power, or carbon dioxide capture and storage technologies (CCS) have to be applied.

Public acceptance of CCS technologies in Europe is relatively low and renewable energies are the preferred greenhouse gas mitigation technologies. This preference of renewable energy production over carbon dioxide sequestration is mainly related to assumed risks involved with transport and storage of carbon dioxide and little knowledge about physical and chemical properties of the gas and associated concerns [138]. However, the actual risk of pipeline transport of carbon dioxide is regarded as similar or lower than that of hydrocarbons and it is expected that appropriately selected and managed geological storage sites will retain more than 99 % of the stored carbon dioxide within 1000 years [54]. Since the use of fossil fuels for energy production is expected to grow in the next decades especially in emerging and developing countries [119], application of CCS technologies could be required if greenhouse gas emissions are to be reduced.

Fossil fuel-fired and particularly coal-fired power stations are important sources of carbon dioxide emissions. Approximately 65 % (4942 in absolute terms) of the worldwide large stationary carbon dioxide sources based on fossil fuels with more than 0.1 Mt CO<sub>2</sub> emissions per year are related to the power sector. These facilities

account for almost 79 % of the carbon dioxide emissions of all large stationary sources. The production of cement accounts for the second largest share with 6.9 % [54]. Fossil fired power stations are therefore especially suited for the application of CCS technologies.

Major purpose of these technologies is to provide highly concentrated carbon dioxide for non-atmospheric disposal. Flue gas with high carbon dioxide concentration requires less energy for compression, transport and storage compared to the treatment of the entire amount of flue gas with low carbon dioxide concentration (typically between 10 to 15 % by vol. for coal-fired systems).

There are three main technologies to capture carbon dioxide emissions from the use of carbon containing fuels: a *pre-combustion* process based on gasification of the fuel, a *post-combustion* process where carbon dioxide is removed from the flue gas by a solvent (e.g. monoethanolamine), and the *oxy-fuel* process. This work is focused on the latter process which achieves a high concentration of carbon dioxide in the flue gas by removing nitrogen from the oxidant gas which is usually air. Nitrogen dilutes carbon dioxide in the flue gas of an air-blown combustor. By removing nitrogen from the system, the flue gas then consists mainly of carbon dioxide, water vapor and minor impurities, e.g. excess oxygen, nitrogen due to leakage air, nitrogen oxides, argon and sulfur species.

### 1.1 Oxy-fuel Process for Coal-Fired Boilers

The idea of burning coal in a mixture of oxygen and recirculated flue gas came up in the 1980s in order to supply carbon dioxide for enhanced oil recovery [1]. Carbon dioxide can be used to facilitate oil recovery by injecting it into oil wells where it decreases viscosity and surface tension of the oil. Later, the process gained more attention for mitigation of carbon dioxide emissions due to increased public concern about climate change.

If air is used as oxidant gas for combustion of fossil fuels, nitrogen from air dilutes carbon dioxide in the flue gas. If the fuel is burned with pure oxygen instead of air, carbon dioxide concentration in the flue gas increases since nitrogen is eliminated from the system. Production of pure oxygen can be done by an air separation unit based on a cryogenic process separating nitrogen and oxygen from air. This cryogenic process is the only available state–of–the–art technology for air separation [63]. It demands a high amount of energy.



Figure 1.1: Basic principle of the coal-fired oxy-fuel process

Combustion of coal with pure oxygen would lead to excessively high temperatures due to a drastically reduced amount of flue gas. Therefore, it is mandatory to recirculate flue gas back into the furnace in order to increase the mass flow of flue gas and by this its heat capacity. The oxidant gas then consists of flue gas and oxygen and combustion temperature can be controlled by adjusting the amount of recirculated flue gas. Combustion of dried lignite (see section 3.1) requires approximately 70 % of the flue gas to be recirculated in order to achieve a similar adiabatic flame temperature as in air-blown combustion [63]. The proportion of the flue gas which is not recirculated can be purified and compressed after removing dust and impurities such as water vapor, sulfur dioxide and nitrogen oxides similar to the conventional process.

Flue gas can be recirculated either upstream or downstream of one of the above mentioned cleaning steps. Flue gas recirculation upstream of the flue gas condenser is sometimes referred to as "wet recirculation" and recirculation downstream of the flue gas condenser is referred to as "dry recirculation" since most of the water vapor is removed. References [25, 136] give an overview on different options of flue gas recirculation. Figure 1.1 shows the basic principle of the coal-fired oxy-fuel process.

Over the last few years there has been plenty of research in order to understand the differences between air-blown and oxy-fuel combustion and the effects on design and operation of coal-fired power stations. Major changes related to oxy-fuel combustion

are:

- other chemo-physical properties of carbon dioxide compared to nitrogen, e.g. higher density and higher heat capacity
- carbon dioxide unlike nitrogen absorbs and emits heat radiation
- reduced flue gas mass flow in the boiler
- drastically reduced flue gas mass flow downstream of the recirculation point
- increased concentration of water vapor and sulfur dioxide if these species are not removed upstream of the recirculation point
- increased concentration of heteroatomic gas species (CO<sub>2</sub>, H<sub>2</sub>O) leads to an increase of gas emissivity
- considerable loss of efficiency mainly due to energy demand for air separation and carbon dioxide compression (estimated in the literature to be between 7 and 13 percentage points compared to conventional power stations [25, 63])

Therefore, research is focused on ignition, combustion, burn out, optimal flue gas recirculation, burner and boiler design,  $NO_x$  formation, sulfur oxides, material issues and heat transfer by radiation and convection. In Europe, first experiments on combustion of coal in a mixture of oxygen and recirculated flue gas were performed in the 1990s by the International Flame Research Foundation (IFRF) [142]. An overview over recent research related to the oxy-fuel process can be found in references [12, 25, 108, 111, 141].

Many of the research results were achieved by operating lab and pilot scale oxyfuel facilities. An overview on different pilot scale studies can be found in [12, 60]. The 30 MW boiler subject to this work is part of the Schwarze Pumpe pilot plant [13, 68, 129, 130] which is one of the largest oxy-fuel facilities worldwide. It features all components required for a full-chain demonstration of the process including air separation unit, boiler, electrostatic precipitator, flue gas desulfurization, flue gas condenser and carbon dioxide purification and compression unit. Therefore, the Schwarze Pumpe test facility was chosen as the ideal plant for the investigation of above mentioned research activities including heat transfer by radiation.

### 1.2 State of the Art

Reference [111] provides a comprehensive overview on coal combustion under oxy– fuel conditions regarding pyrolysis, char combustion, burner design,  $NO_x$  emissions, sulphur chemistry, slagging, fouling and corrosion. It includes also a review on recent investigations of radiative heat transfer and its modeling in oxy–fuel combustion.

Unlike nitrogen, carbon dioxide absorbs and emits heat radiation due to its heteroatomic molecular structure. Since the concentration of carbon dioxide in the flue gas of oxy-fuel furnaces is higher compared to conventional furnaces, flue gas emissivity and absorptivity are expected to increase. Emissivity describes the ability of substances to emit heat radiation in comparison with a black body. It is an intensive quantity. The ability to absorb heat radiation is often referred to as absorptivity. Emissivity and absorptivity of gaseous species are functions of wavelength. Gases emit and absorb radiation only at certain wavelengths and are transparent at others.

Water vapor is another radiating gas species present in the flue gas of coal-fired boilers. Its concentration in the flue gas depends mainly on the water and hydrogen content of the fuel. Due to flue gas recirculation and lower flue gas volume, water vapor concentration is higher in the flue gas of oxy-fuel boilers. The concentration is particular high in the case of "wet recirculation" of flue gas downstream of the electrostatic precipitator [136].

The effect of increased partial pressures of radiating species on heat radiation in coalfired boilers and its modeling is unclear and needs to be evaluated and quantified. Emissivity of the gas-particle-dispersion in a furnace is often dominated by particle emission. There are four main kinds of solid particles present in a coal-fired furnace: coal, char, ash and soot. Unlike carbon dioxide and water vapor, solid particles are generally assumed to radiate continuously over the spectrum. Therefore, increase in gas emissivity might have only minor effects on the overall emissivity of gasparticle-mixtures.

Taking the above into account, calculation of heat transfer by radiation in oxyfuel atmosphere requires two steps: an accuracy check of the applied gas emissivity model at higher partial pressures of radiating gas species (i.e., carbon dioxide and water vapor) and evaluation of increased gas emissivity on the overall gas-particle emissivity. Once the optical properties are determined, heat transfer by radiation can be calculated. Since particle radiation dominates in coal-fired furnaces [111], influence of gas emissivity on radiative heat transfer might be small.

There are different kinds of gas optical properties models which need to be validated for oxy-fuel combustion [82, 111]. They differ in accuracy and computational effort. In engineering calculations, it is often desirable to use simplified gas optical properties models, so-called "gray models". These models do not account for spectral dependency of gas emissivity and instead provide gray or total emissivities. Thus, changes of gas emissivity due to increase in partial pressure of radiating gas species has to be evaluated against the background of general accuracy of the applied emissivity model. A frequently used emissivity model is the weighted sum of gray gases model (WSGGM) with three gray gases and one transparent gas (WSGG3+1) by Smith [122]; see section 2.2.2.

In case of gray calculation, spectrally independent absorption and emission coefficients are required as extensive quantities for the solution of the function that describes heat transfer by radiation. They are calculated from gas emissivity and absorptivity, respectively. There are different methods to calculate spectrally averaged gas emission and absorption coefficients. These methods are valid only in certain optical path length ranges. Therefore, derivation of spectrally averaged gas absorption and emission coefficients for gray solutions has to be evaluated regarding increased concentrations of radiating gas species in oxy-fuel fired furnaces.

There are other very efficient absorptivity and emissivity correlations which provide higher accuracy, e.g. the wide band correlated–k model (WBCK) [128]. However, gray solutions offer the lowest computational effort since their spectral resolution is lowest: the radiation problem only has to be solved once and not multiple times for certain wavelengths, wavelength intervals or spectral blocks. The higher computational effort of models with higher spectral resolution is regarded as not justified in case of oxy–fuel combustion systems [82]. The influence of gas absorption and gas emission coefficient on radiative heat transfer is expected to be low and gray solution of radiative transfer in oxy–fuel furnaces might give satisfactory results just like for air–blown combustion.

In oxy-fuel furnaces, gas temperatures can be higher or lower than in air-blown furnaces depending on the amount of recirculated flue gas. Radiation intensity, however, is a function of temperature. Therefore, gas temperatures strongly influence radiation processes. Among other things, gas temperatures are in turn results of radiative processes. The influence of gas optical properties on radiative heat transfer has to be evaluated against this background. Therefore, the influence of increased carbon dioxide and water vapor concentration has to be compared with the influence of gas temperature variation by univariant analysis which means that sensitivity of radiative heat fluxes towards only one of the parameters is investigated.

### 1.3 Scope of Work

This work includes the evaluation of modeled gas absorption coefficients and underlying gas emissivities at elevated concentrations of radiating gas species and an accuracy check of the applied models. Scheffknecht et al. [111] pointed out that, besides comparisons against benchmark calculations and measurements of propane flames, validation of suitable emissivity models against experimental data of pulverized coal combustion under oxy-fuel conditions is required. Therefore, radiative heat transfer in a pilot-scale 30 MW oxy-fuel furnace is calculated. The results are then compared with measured values of incident radiative wall heat fluxes.

The size of the regarded combustion chamber allows to obtain representative results with respect to optical path length. The average path length in laboratory scale test furnaces is small in comparison to commercial boilers which hinders the analysis of radiative property models [111] and the significance of measurement and simulation results is limited. The average optical path length of the regarded pilot–scale furnace, however, is large enough to provide meaningful results. To the author's knowledge, there is no published comparison of measured and simulated heat radiation results for an oxy–fuel system of this size available at present. Kangwanpongpan in his thesis [60] provides a comprehensive overview on investigated laboratory and semi– industrial scale oxy–fuel systems.

Heat transfer by radiation is calculated by integrating a transport equation of radiative intensity over space coordinates, solid angle and wavelength. Since this can be very complex, some simplifications are introduced in engineering calculations in order to reduce computation time. One fundamental assumption is that flue gas emissivity and absorptivity are equal<sup>1</sup>. Optical properties of gaseous combustion products depending on wavelength are regarded as gray (i.e., independent of wavelength). This drastically simplifies calculation of radiative heat transfer. Gray modeling seems to be favorable also for oxy-fuel combustion systems [82]. However, accurate models to predict wavelength averaged optical properties are required for this purpose.

<sup>&</sup>lt;sup>1</sup>For the sake of semantic convenience, the term "emissivity" is used synonymously for both, emissivity and absorptivity in the following if this fundamental simplification of "gray" modeling applies.

State-of-the-art models were developed for combustion calculations but are mostly limited to low concentrations of carbon dioxide and water vapor. Therefore, one of these models is checked for accuracy and "recalibrated" for higher concentrations of radiating species. The adapted gray emissivity correlation is then compared to recently developed models from the literature. Flue gas emissivity of the regarded combustion system is then calculated with the help of the recalibrated model in order to determine the increase in gas emissivity due to oxy-fuel combustion. After that, a grav absorption coefficient<sup>2</sup> is derived from the intensive gas optical properties. It is worth to mention that a lot of recent work has been done on calculating flue gas emissivity as accurate as possible also under oxy-fuel conditions with elevated concentrations of carbon dioxide and water vapor. However, to the author's knowledge, there is no evaluation on how spectrally averaged absorption coefficients can be calculated under these conditions. Therefore, two different methods to calculate gray, spectrally averaged absorption coefficients are compared to each other and investigated in detail. Finally, radiative heat transfer to the furnace walls is calculated and compared to measured values. The influence of increased gas emissivity on radiative wall fluxes is evaluated with respect to different gas temperatures.

Modeling of radiative heat transfer in the furnace is *non-iterative* based on predefined temperatures and gas species concentrations derived from measurements. This is done in order to focus on calculation of radiative transfer, to exclude influences from flow and chemical reaction models and to avoid computational expensive, iterative solutions. The heat transfer problem is solved by Monte Carlo simulation following a large number of beams on their way through the furnace. The simulation is based on random numbers for the determination of the beam's start position, start angle, reflection angle and certain events like absorption and scattering. Since coal, ash, and soot particles are present in the flue gas of coal-fired steam generators, simplified gray approaches are used to calculate optical properties of these particles.

If the gas temperature distribution in the furnace is not known from measurements, it has to be calculated from an *iterative* solution of radiative heat transfer. This means that temperatures and radiative heat transfer are consecutively calculated until the solution converges towards stable values. Iterative solution is relevant for heat transfer calculation if information on temperature distribution is not available which is usually the case in engineering applications. Therefore, the adapted gas emissivity correlation is implemented as user–defined function in the Computational

<sup>&</sup>lt;sup>2</sup>For the sake of semantic simplicity, the term "absorption coefficient" is used synonymously hereafter for both, absorption and emission coefficient if gray simplifications apply.

Iterative solution	$Non-iterative \ solution$
Purpose is the prediction of combustion processes	Purpose is the calibration of optical properties models
Simulation tool FLUENT, chapter 6	Monte Carlo ray tracing, chapter 4
Calculation of temperatures	Temperatures defined a posteriori
Calculation of radiative heat flux at the furnace walls	Calculation of radiative heat flux at the furnace walls
Optical properties influence gas temperature and radiative heat flux	Optical properties only influence radia- tive heat flux
Computational effort is high	Computational effort is low

 
 Table 1.1: Main attributes of iterative and non-iterative solution approach of radiative transfer

Fluid Dynamics (CFD) tool FLUENT. This is done in order to understand the significance of gas emissivity and gas absorption coefficient in case of iterative solution of radiative transfer.

The *non-iterative* solution using predefined temperatures serves rather as calibration of optical property models. Gas temperatures are derived a posteriori from measurements. In this case, gas optical properties influence only the heat flux results. Accurate optical properties models will result in good agreement between measured and calculated heat fluxes. Table 1.1 shows main attributes of both approaches.

# 2 Fundamentals of Radiative Heat Transfer

The quantity describing the energy transported by radiation at a certain wavelength into a certain direction  $\vec{s}$  is the spectral radiative intensity  $I_{\nu}$ . It is defined as the "radiation energy per unit time per unit area per unit solid angle normal to the area" [49] within an infinitesimal wavenumber interval  $d\nu$ :

$$I_{\nu} = \frac{d\dot{Q}}{dA\,d\Omega\,d\nu}.\tag{2.1}$$

The spectral radiative intensity into a certain direction does not change along a path s in the vacuum. In radiatively participating media, however, the intensity does change due to absorption, emission and scattering.

Radiation energy passing through participating medium is absorbed according to the absorption properties of the medium. The amount of absorption is directly proportional to the magnitude of incoming intensity and path length s [92] and can be written as:

$$dI_{\nu} = -\kappa_{\nu,abs} I_{\nu} ds. \tag{2.2}$$

The proportionality constant  $\kappa_{\nu,abs}$  is the spectral absorption coefficient of the medium describing its absorption properties. In combustion systems, these properties depend on the concentration of participating flue gas species, e.g. carbon dioxide and water vapor. The absolute amount of attenuation of radiative intensity by absorption can be found according to Lambert–Beer's law [49]

$$I = I_0 \exp\left[-\kappa L\right]. \tag{2.3}$$

It describes the logarithmic dependence of attenuation of radiative intensity from the attenuation coefficient  $\kappa$  multiplied by path length L. Integration of equation 2.2 along path s gives

$$I_{\nu,s} = I_{\nu,0} \exp\left[-\int_{0}^{s} \kappa_{\nu,abs} \, ds\right]$$
(2.4)

(a) 
$$I_{b,\nu}(T)$$
 (b)  $F(\nu,T)$ 



Figure 2.1: Spectral black body intensity  $I_{b,\nu}(T)$  (a) and fractional black body function  $F(\nu, T)$  (b) as functions of the wavenumber  $\nu$  at temperatures 1000, 1200 and 1400 K

where  $I_{\nu,0}$  is the incident radiative intensity at s = 0. In scattering medium, incident radiation is scattered according to the medium's properties and intensity in direction  $\vec{s}$  is attenuated:

$$dI_{\nu} = -\sigma_{\nu,s}I_{\nu}ds. \qquad (2.5)$$

Integration along s gives:

$$I_{\nu,s} = I_{\nu,0} \exp\left[-\int_{0}^{s} \sigma_{\nu,s} \, ds\right].$$

$$(2.6)$$

The total amount of attenuation of incident radiation intensity is described by the spectral extinction coefficient  $\beta_{\nu}$  which is the sum of spectral absorption coefficient  $\kappa_{\nu,abs}$  and spectral, directional scattering coefficient  $\sigma_{\nu,s}$ .

Incident intensity is increased due to emission and in–scattering of radiative energy. Emission of energy into the regarded direction along path s depends on optical and thermodynamic properties of the medium. The increase of radiative intensity can be described by

$$dI_{\nu} = \kappa_{\nu,em} I_{b,\nu}(T) ds \tag{2.7}$$

where  $I_{b,\nu}(T)$  is the spectral black body intensity of the medium with temperature T defined as [92]

$$I_{b,\nu}(T) = \frac{C_1 \nu^3}{e^{C_2 \nu/T} - 1}$$
(2.8)

where  $\nu$  is the wavenumber in  $[m^{-1}]$ . The coefficient  $\kappa_{\nu,em}$  is the spectral emission coefficient. Figure 2.1(a) shows the spectral black body intensity  $I_{b,\nu}(T)$  as function of wavenumber for T = 1000, 1200 and 1400 K. The Planck radiation constants  $C_1$  and  $C_2$  are [92]:

$$C_1 = 2\pi hc^2$$
$$C_2 = hc/k_B$$

with h being the Planck constant, c the speed of light and  $k_B$  the Boltzmann constant. Integration over all wavelengths gives the total black body intensity [92]

$$I_b(T) = \int_{\nu=0}^{\infty} I_{b,\nu}(T) d\nu = \frac{\sigma_c}{\pi} T^4$$
(2.9)

where  $\sigma_c$  is the Stefan–Boltzmann constant ( $\sigma_c = 5.670 \cdot 10^{-8} \text{ Wm}^{-2} \text{K}^{-4}$ ). The fractional function of black body radiation  $F(\lambda, T)$  with  $\lambda = 1/\nu$  at temperature T is defined as [76]:

$$F(\lambda,T) = \frac{\int\limits_{0}^{\lambda} E_{b,\lambda} d\lambda}{\int\limits_{0}^{\infty} E_{b,\lambda} d\lambda}$$
(2.10)

where  $E_{b,\lambda}$  is the spectral black body emissive power of a volume,  $4\pi \cdot I_{b,\lambda}(T)$  if intensity is isotropic [49]. Approximative functions of  $F(\nu, T)$  were published for easy computational application. For  $\chi = C_2 \nu/T \ge 2$  the relation

$$F(\nu,T) = \frac{15}{\pi^4} \sum_{n=1}^{4} \left[ \frac{e^{-n\chi}}{n} \left( \chi^3 + \frac{3\chi^2}{n} + \frac{6\chi}{n^2} + \frac{6}{n^3} \right) \right]$$
(2.11)

is used [16]. For  $\chi < 2$  the approximation

$$F(\nu,T) = 1 - \frac{15}{\pi^4} \chi^3 \left( \frac{1}{3} - \frac{\chi}{8} + \frac{\chi^2}{60} - \frac{\chi^4}{5040} + \frac{\chi^6}{272160} - \frac{\chi^8}{13305600} \right)$$
(2.12)

is applied [118]. Figure 2.1(b) shows the fractional black body function  $F(\nu, T)$  for T = 1000, 1200 and 1400 K as function of wavenumber.

In-scattering of radiation energy over the whole solid angle of  $4\pi$  is described by



Figure 2.2: Scheme of the radiative transfer equation according to [128]

[92]

$$dI_{\nu} = \frac{\sigma_{\nu,s}}{4\pi} \int_{4\pi} I_{\nu}(\vec{s_i}) \Phi_{\nu}(\vec{s_i} \to \vec{s}) d\Omega_i \, ds \tag{2.13}$$

where  $\Phi_{\nu}(\vec{s_i} \to \vec{s})$  is the scattering phase function. For isotropic in–scattering the phase function becomes  $\Phi_{\nu}(\vec{s_i} \to \vec{s}) \equiv 1$  [92] which means that in–scattering is independent of direction.

#### 2.1 Radiative Transfer Equation

Change in radiation intensity by emission, absorption and scattering along a path s in direction  $\vec{s}$  can be found from summation of equations 2.2, 2.5, 2.7 and 2.13:

$$\frac{dI_{\nu}}{ds} = \kappa_{\nu,em}I_{b,\nu} - \kappa_{\nu,abs}I_{\nu} - \sigma_{\nu}I_{\nu} + \frac{\sigma_{\nu,s}}{4\pi} \int_{4\pi} I_{\nu}(\vec{s_i})\Phi_{\nu}(\vec{s_i} \to \vec{s})d\Omega_i \qquad (2.14)$$

where  $\kappa_{\nu,em}$  is the spectral emission coefficient,  $\kappa_{\nu,abs}$  is the spectral absorption coefficient and  $\sigma_{\nu}$  is the spectral scattering coefficient. This statement of energy conservation for a monochromatic intensity along way *s* in direction of  $\vec{s}$  is called radiative transfer equation (RTE) [76, 139] in Lagrangian coordinates and is illustrated in figure 2.2. Reference [139] provides detailed derivation of the RTE and its formulation in Eulerian coordinates.

The derivation of the RTE is based on the assumption that the regarded medium is in local thermodynamic equilibrium (LTE) [76]. According to *Kirchhoff's law*, emissivity and absorptivity are equal in LTE. Thermodynamic equilibrium means that the thermodynamic properties of the medium are constant over space and time and there are no net flows of energy or matter from or into the medium. According to [76], LTE implies three conditions:

- The regarded system can be divided into a number of control volumes within which thermodynamic properties such as temperature, internal energy, density and pressure are constant.
- Fluctuation of thermodynamic properties is small compared to their average value.
- Time scale of radiative processes is infinitely fast compared to the rate of change of thermodynamic properties.

The concept of LTE in radiation modeling is explained in detail in [76]. Derivation of Kirchhoff's law for radiating gas volumes can be found in [49].

If scattering is neglected, the RTE simplifies to

$$\frac{dI_{\nu}}{ds} = \kappa_{\nu,em} I_{b,\nu} - \kappa_{\nu,abs} I_{\nu}.$$
(2.15)

Scattering, however, can only be neglected if small soot particles are the only dispersed particles present in the flue gas [76]. This might be the case for oil or gas flames but not for combustion of solid, pulverized fuels. Therefore, scattering must be considered in the calculation of heat transfer in coal-fired furnaces.

#### 2.1.1 Gray Formulation

If emission coefficient  $\kappa_{\nu,em}$  and absorption coefficient  $\kappa_{\nu,abs}$  in equation 2.15 are spectrally averaged, the semi–gray RTE [76, 137] can be written as

$$\frac{dI}{ds} = \overline{\kappa}_{em} I_b - \overline{\kappa}_{abs} I. \tag{2.16}$$

Regarding absorption and emission coefficient as equal  $\overline{\kappa}_{em} = \overline{\kappa}_{abs}$  yields the gray formulation of the RTE [76]

$$\frac{dI}{ds} = \overline{\kappa} \left( I_b - I \right) \tag{2.17}$$

where  $\overline{\kappa}$  is the spectrally independent, gray absorption and emission coefficient<sup>1</sup>. Equation 2.17 is commonly used in engineering calculations to compute radiative heat transfer. It is shown by [137] that the RTE with a common coefficient  $\overline{\kappa}$  for absorption and emission becomes meaningless if temperature gradients occur (i.e., inhomogeneous temperature distribution). Viskanta and Mengüç [139] state that the implied equality of absorption and emission coefficient is justified as long as there are no large temperature gradients in the medium and if soot concentration is high<sup>2</sup> so that spectral absorption and emission coefficient of the gas-particle-dispersion are only weak functions of wavelength.

A number of gray optical property models has been published where gray emissivity of gaseous combustion products is calculated from polynomials [45, 79, 91, 110], weighted sums of gray gases [7, 21, 33, 34, 49, 58, 122, 126, 133, 134] or other expressions [91]. Based on modeled emissivity, the gray absorption coefficient  $\overline{\kappa}$  that is required in equation 2.17 can be calculated.<sup>3</sup> More details on how to calculate gray absorption coefficients from gas emissivity are given in section 2.2.4.

#### 2.1.2 Solution of the Radiative Transfer Equation

An overview on solution methods for the integro-differential RTE was presented by Viskanta and Mengüç [139] and by Carvalho and Farias [15]. Examples for exact solutions of the RTE can be found in [22, 23, 113, 115]. However, exact solutions are often not practical for engineering calculations where simplified approximations are required [15].

Hottel and Cohen [48] developed the *zone method* to predict radiative heat transfer in furnaces. The furnace is subdivided into volume and surface zones. Temperature and optical properties are assumed to be constant within each zone. View factors are calculated for each gas–gas, gas–surface and surface–surface exchange and net exchange for each pair of zones is then determined. View factors for cylindrical enclosures are presented in [49]. The zone method, however, requires high computational effort since for every zone radiative exchange with every other zone has to be calculated. Results of zone method application were presented in [58, 59, 67] and as benchmark results in [114].

<sup>&</sup>lt;sup>1</sup>For semantic convenience referred to as *absorption coefficient* in the following.

<sup>&</sup>lt;sup>2</sup>Solid particles unlike gases emit and absorb heat radiation continuously across the spectrum so that wavelength dependency of optical properties becomes low in gas-particle mixtures with sufficient concentration of solid matter.

<sup>&</sup>lt;sup>3</sup>A gray absorption coefficient (extensive quantity) is required rather than emissivity (intensive quantity) for the differential solution of radiative transfer [139].

Ray tracing methods, such as the *Monte Carlo method*, are based on following a number of random beams through the regarded domain. The Monte Carlo method is described in more detail in section 2.1.3 since it is applied in chapter 4. It is a highly accurate method and the solution "should converge to the exact solution" if the number of emitted beams increases to infinity [15]. The *discrete transfer method* [86, 116] divides the domain into volume elements with constant temperatures and optical properties with beams starting from surface elements. The hemisphere over the center of a surface element is subdivided into a certain number of fixed directions. The RTE is then solved for an emitted beam in each direction until it arrives at another surface element. The method was applied in [14, 19] and extended for multi-dimensional, non-gray systems in [85].

There are two other solution methods which are commonly applied in radiative heat transfer calculations [92]: the spherical harmonics approximation and the discrete ordinates method. Both methods transform the RTE into a system of partial differential equations. The discrete ordinates method [37–39] or its variation, the finite volume method [20], discretizes the whole solid angle of  $4\pi$  into a number of directions within which the intensity is assumed to be constant. The RTE is then solved for each direction. In the spherical harmonics approximation, directional dependence of the RTE is approximated by truncated series expressions [92].

#### 2.1.3 The Monte Carlo Method

The Monte Carlo method is a statistical solution of the RTE based on random numbers. The name refers to the administrative area of the principality of Monaco famous for its casinos. Fleck [40] and Howell and Perlmutter [52, 53] primarily applied Monte Carlo simulation to radiative heat transfer problems. Howell gives a simple example of application to thermal radiation in his technical memorandum [50]. In the following, a similar example is given in order to explain the RTE solution procedure in section 4.

The Monte Carlo method follows a number of beams on their way through the regarded domain. Starting point, direction, wavelength, absorption and scattering of each emitted beam can be determined by probability functions of the form

$$R = \int_{\xi} P(\xi) d\xi.$$
 (2.18)
(a) Probability function

(b) Cumulative distribution function



Figure 2.3: The (a) cosine probability function  $p(\lambda)$  and (b) the probability density function  $P(\lambda)$  (dotted line) with the corresponding cumulative distribution function  $R_{\lambda}$  (solid line)

where R is a random number in the range 0 < R < 1. Random numbers are uniformly distributed within that range. Equation 2.18 is called fundamental theorem of Monte Carlo [51] or cumulative distribution function [50]. Values for  $\xi$  are obtained by randomly choosing R and solving equation 2.18. The distribution of random values corresponds with the physical process to be described.

As an example, distribution of emissivity  $\epsilon(\lambda)$  is described by equation

$$\epsilon(\lambda) = 0.2\cos(\lambda) + 0.2 \tag{2.19}$$

within the wavelength interval  $0 \le \lambda \le 30 \cdot 10^{-6}$  m, see figure 2.3(a). The probability density function is then defined as [50]

$$P(\lambda) = \frac{\epsilon(\lambda)}{\int\limits_{0}^{30} \epsilon(\lambda) d\lambda}$$
(2.20)

giving for this example

$$P(\lambda) = \frac{\cos(\lambda) + 1}{\sin(30) + 30}$$

Figure 2.3(b) shows the probability density function (dotted line). The cumulative distribution function is then calculated according to equation 2.18. For this example

the cumulative distribution functions reads

$$R_{\lambda} = \int_{0}^{30} P(\lambda) d\lambda = \frac{\sin(\lambda) + \lambda}{\sin(30) + 30}.$$

For each  $R_{\lambda}$  being a random number between 0 and 1, the cumulative distribution function yields a wavelength within the respective wavelength interval. After choosing a large number of random events, the resulting wavelength distribution satisfies the underlying physical relation. In this manner, the physical process can be described by a number of random events. However, this number has to be large enough to produce statistically relevant results. In section 4, the Monte Carlo method is applied to simulate radiative processes in the regarded furnace.

# 2.2 Gas Emissivity Modeling

In order to calculate heat transfer by radiation through radiatively participating medium, radiative properties of the medium must be known. Radiation of gas molecules is limited to certain wavenumber intervals. The distribution of these intervals, also called wavenumber bands, is different for each gas depending on its molecular structure. Homonuclear gases like nitrogen and oxygen are non-radiative in the infrared while heteronuclear gases like carbon dioxide and water vapor absorb and emit radiation within characteristic spectral regions in the infrared.

In order to reduce computational effort, the fundamental character of gases to radiate only within certain wavenumber bands is often neglected. The ability of a gas to emit heat radiation independent of wavelength is called gray or sometimes total emissivity. It can be used to calculate<sup>4</sup> a spectrally independent, gray absorption coefficient<sup>5</sup>. The number of required RTE solutions  $N_{RTE}$  depends on spectral and spatial discretization:

$$N_{RTE} = N_{\nu} \cdot N_{\vec{s}} \tag{2.21}$$

with  $N_{\nu}$  being the number of wavelength intervals and  $N_{\vec{s}}$  the number of directions [82]. Finding appropriate expressions for gray emissivity leads to  $N_{\nu} = 1$  since the wavenumber dependency is eliminated. This reduces the required number of RTE solutions drastically and is the major advantage of this approach.

 $^4$ Section 2.2.4 shows how gray absorption coefficients can be determined from gray gas emissivity.

<sup>&</sup>lt;sup>5</sup>The gray absorption coefficient  $\overline{\kappa}$  is required in the gray RTE formulation, equation 2.17.

# 2.2.1 Overview on Emissivity Models

There are several models to calculate radiative characteristics of gases and gas mixtures. Figure 2.4 gives an overview on different emissivity correlations. So–called line–by–line (LBL) models provide the highest spectral resolution and, thus, require the highest computational effort since a large amount of spectral data has to be calculated [75]. LBL models are based on large databases [102, 105, 112]. Due to their high computational effort, they are mainly used as benchmark models in engineering sciences.

Band models resolve the infrared spectrum into several wavenumber intervals. Narrow band models (NBM), like the statistical narrow band model (SNBM) [83] or as in RADCAL [44], are described in detail by Ludwig et al. [88]. Pierrot, Soufiani and Taine [125] studied the accuracy of band and global models in comparison with an LBL approach [98]. Parameters have been published by several authors [46, 96, 97, 125]. NBM provide most accurate radiative properties of combustion gases but similar to LBL models they are computationally too expensive to be applied in engineering software tools for flame calculations [75] since the spectrum is resolved into hundreds of bands and the RTE has to be solved for each of them.

Taine and Soufiani [131] developed the correlated–k (CK) model for narrow bands based on a cumulative distribution function. The correlated–k model with fictitious gases (CKFG) is an improvement of the CK model for strong non–isothermal media [103]. An efficient wide band correlated–k model was developed in [128]. However, k–distribution methods are not described in detail here since the increase in computational effort compared to gray solutions is rather not justified [82].

The exponential wide band model (EWBM) requires less computational effort than NBM. The EWBM concentrates on a small number of wavenumber intervals in the near infrared and infrared [75]. The model has been developed by Edwards and coworkers [30, 31]. The EWBM is described in more detail in section 2.2.3 as it serves as benchmark model in this work. Correlated–k based wide band models are described in the literature [80, 89, 128].

Besides the models described before, there are several global models for prediction of emissivity of gases or gas mixtures. Global models are "gray" (i.e., spectrally independent) expressions of gas optical properties. Hottel and Sarofim [49] provided emissivity charts for carbon dioxide and water vapor for engineering calculations. They also introduced the weighted sum of gray gases model (WSGGM) (see section 2.2.2). Other expressions for total emissivities have been presented [45, 79, 91, 110].



Figure 2.4: Overview on different emissivity correlations

The model of Leckner [79] is regarded as most general [76] since it takes into account total pressure and partial pressures of carbon dioxide and water vapor. Therefore, this model is basically applicable also for flue gas mixtures of oxy-fuel combustion. However, the model is valid only for carbon dioxide partial pressure path lengths  $p_{CO_2}L < 0.1 \cdot 10^6$  mPa [76] which limits its application to rather small optical path lengths (see section 4.3.3).

An important improvement of the WSGGM is the spectral line–based weighted sum of gray gases model (SLW) of Denison and Webb [27–29]. The absorption distribution function (ADF) model and the absorption distribution function model with fictitious gases (ADFFG) [99] are recently developed global models. However, the WSGGM continues to be the most important global model in engineering applications [99].

#### 2.2.2 Weighted Sum of Gray Gases Model (WSGGM)

In engineering calculations, it is often postulated that radiative media are gray and attenuate radiation according to Lambert–Beer's law, equation 2.3. Real gas, however, absorbs radiation only at characteristic wavelengths. The WSGGM expresses spectral characteristics of real gas as required for gray calculations. The model was introduced by Hottel and Sarofim for the zone method for calculating radiative transfer in furnaces [49] with black walls filled with non–scattering media. However, the model is also applicable for gray walls and gray scattering under certain conditions [144].

Gray gas emissivity can be calculated from the model<sup>6</sup>. Monochromatic emissivity and absorptivity are the same according to Kirchhoff's law [49]

$$\epsilon_{\nu} = \alpha_{\nu} \tag{2.22}$$

and from equation 2.3 emissivity and absorptivity relate to absorption coefficient according to

$$\epsilon_{\nu} = \alpha_{\nu} = 1 - \exp[-\kappa_{\nu}L]. \tag{2.23}$$

Section 2.2.4 shows the calculation of gas absorption coefficients from gas emissivity under gray conditions.

<sup>&</sup>lt;sup>6</sup>More precisely, the model gives the gray *standard emissivity* of a gas, the ratio of incident radiation onto an infinitesimal surface "from a narrow pencil of rays of length L to incident radiation [...] from a blackbody at gas temperature" [49].

The WSGGM calculates gas emissivity as weighted sum of u gray gas terms

$$\epsilon_g = \sum_u a_{\epsilon,u} \left[ 1 - e^{-\kappa_u pL} \right] \tag{2.24}$$

where  $a_{\epsilon,u}$  are weighting factors and pL is the pressure path length of the gas mixture. For large numbers of u,  $a_{\epsilon,u}$  could be considered as fraction of black body energy in spectral regions with absorption coefficient  $\kappa_u$  [49]. One term of the series is often assigned a value of  $\kappa_u = 0$  in order to consider spectral regions where the gas mixture is transparent. All values of  $a_{\epsilon,u}$  must be positive and

$$\sum_{u} a_{\epsilon,u} = 1. \tag{2.25}$$

There have been numerous publications of coefficients or coefficient expressions for the WSGGM [7, 21, 33, 34, 49, 58, 122, 126, 133, 134]. Lallemant [76] found that the model of Smith et al. [122] is in best agreement with the EWBM over a wide range of tested pressure path lengths and temperatures. Thus, this emissivity correlation is often used in engineering software tools [41, 72].

Smith and coworkers use a correlation with three gray gases and one clear gas

$$\epsilon_g = \sum_{u=0}^3 a_{\epsilon,u} \left[ 1 - e^{-\kappa_u pL} \right]$$
(2.26)

with  $\kappa_0 = 0$  and

$$a_{\epsilon,0} = 1 - \sum_{u=1}^{3} a_{\epsilon,u} \tag{2.27}$$

and the other weighting factors  $a_{\epsilon,u}$  represented by temperature polynomials of degree three to allow for variation in gas temperature:

$$a_{\epsilon,u} = \sum_{v=1}^{3} b_{\epsilon,u,v} T^{v-1}.$$
 (2.28)

The series is then fitted to benchmark data generated from the EWBM using Fletcher–Powell technique [73] for atmospheric pressure, gas temperatures from 600 to 2400 K and 50 K intervals, and a pressure path length range of 101.33 to  $1.0133 \cdot 10^6$  mPa. Coefficients are given in table 4.6 in section 4.3.2.

The model of Smith and coworkers, however, was developed for stoichiometric combustion of oil and methane with air: the molar fraction of carbon dioxide in the flue gas being  $\psi_{CO_2} = 0.1$  and ratios of water vapor and carbon dioxide mole fraction  $\psi_{H_2O}/\psi_{CO_2} = \Psi = 1$  and  $\Psi = 2$ . Khare and Wall [65] show that both, increased partial pressure of radiating species and mole fraction ratios  $\Psi < 1$  necessitate new or additional WSGG parameters for accurate prediction of optical properties of gas mixtures in oxy-fuel systems.

The limitation of published WSGG model parameters to ratios  $\Psi$  being one or two has been addressed since the ratio may be significantly different in near burner regions [75]. Krishnamoorthy and coworkers [71, 72] found that application of WSGG parameters according to [122] outside of their  $\Psi$ -range to coal-fired oxy-fuel combustion results in significant error compared to EWBM based modeling.

Recently, Johansson and coworkers [56, 57] published four gray gases plus one clear gas approaches where both weighting factors and absorption coefficients are represented by linear functions or polynomials of  $\Psi$  in the range  $0.125 \leq \Psi \leq 2$ . Yin et al. [143] published new WSGG coefficients for fixed mole fraction ratios  $\Psi$ . By this, they allow for different compositions of oxy-fuel flue gas.

Recently, Kangwanpongpan and coworkers [61] developed coefficients for polynomial functions of the molar ratio  $\Psi$  representing WSGG absorption coefficients and polynomial functions of temperature to calculate WSGGM weighting factors. Their model is based on line–by–line calculations from HITEMP 2010 database [105, 106].

Marzouk and Huckaby [90] give an overview on recent work done to model gas emissivities in oxy–fuel combustion systems. They applied recently published WSGGM approaches to radiative heat transfer simulation in a large, rectangular enclosure filled with different mixtures of carbon dioxide and water vapor. They found significant improvement of radiative heat flux prediction by using WSGGM parameters suited for oxy–fuel combustion<sup>7</sup> instead of weights and absorption coefficient published by Smith et al. [122].

# 2.2.3 Exponential Wide Band Model (EWBM)

The EWBM serves as benchmark model in order to find new weights and absorption coefficients for the WSGG model. While NBM are based on large databases with average spectral properties tabulated for wavenumber intervals between 5 and 25 cm<sup>-1</sup> [88], wide band models approximate these properties by analytical expressions. The

 $<sup>^7\,\</sup>mathrm{They}$  tested five different sets of WSGGM parameters published by different authors.

EWBM approximates the band absorption coefficient  $\overline{\kappa}_{\nu}$  by an exponential function [31].

The model considers five bands for water vapor (with band centers  $\nu_{C,h,j}$  at wavenumbers 7250, 5350, 3760, 1600 and 140 cm<sup>-1</sup>) and six bands for carbon dioxide (with band centers  $\nu_{C,c,j}$  at wavenumbers 5200, 3660, 2410, 1060, 960 and 667 cm<sup>-1</sup>). A four-region approximate expression for total absorptance  $A_{i,j}$  and transmissivity  $\tau_{i,j}$  of the *j*th band of the *i*th gas in the gas mixture was proposed by Edwards and Menard [31]:

In the *linear* region with  $\tau_{H,i,j} \leq 1$  and  $\tau_{H,i,j} \leq \eta_{i,j}$ 

$$A_{i,j} = \omega \cdot \tau_{H,i,j}$$
$$\tau_{i,j} = 0.9,$$

in the square root region with  $\eta \leq \tau_{H,i,j} \leq 1/\eta$  and  $\eta \leq 1$ 

$$\begin{aligned} A_{i,j} &= \omega_{i,j} \left( (4 \cdot \eta_{i,j} \cdot \tau_{H,i,j})^{1/2} - \eta_{i,j} \right) \\ \tau_{i,j} &= 0.5 \left( 1 + \frac{\omega_{i,j} \cdot \eta_{i,j}}{A_{i,j}} \right), \end{aligned}$$

in the log-root region with  $\tau_{H,i,j} \geq 1/\eta$  and  $\eta_{i,j} \leq 1$ 

$$A_{i,j} = \omega_{i,j} \left( \ln(\tau_{H,i,j} \cdot \eta_{i,j}) + 2 - \eta_{i,j} \right)$$
  
$$\tau_{i,j} = \frac{\omega_{i,j}}{A_{i,j}}$$

and in the logarithmic region with  $\tau_{H,i,j} > 1$  and  $\eta_{i,j} > 1$ 

$$A_{i,j} = \omega_{i,j} \left( \ln(\tau_{H,i,j}) + 1 \right)$$
  
$$\tau_{i,j} = \frac{\omega_{i,j}}{A_{i,j}}$$

where  $\tau_{H,i,j}$  is the optical depth at band head or band center defined as

$$\tau_{H,i,j} = \frac{\alpha_{i,j} \cdot \rho_i \cdot L}{\omega_{i,j}} \tag{2.29}$$

with  $\rho_i$  being the density of gas i, L being the path length and  $\alpha_{i,j}$  the integrated

band intensity. The variable  $\eta_{i,j}$  is the line width to spacing parameter defined as

$$\eta_{i,j} = \beta_{i,j} \cdot P_{eq,i,j}. \tag{2.30}$$

 $P_{eq,i,j}$  is the dimensionless equivalent broadening pressure calculated from

$$P_{eq,i,j} = \left(\frac{P_t}{P_0}(1+\psi_i(b_i-1))\right)^{n_{i,j}}$$
(2.31)

where  $P_t$  is the total pressure of the gas,  $P_0$  is the reference pressure,  $\psi_i$  the mole fraction of gas *i* and  $b_i$  and  $n_{i,j}$  fitting parameters given in [76]. Parameter  $\beta_{i,j}$  is calculated from a temperature function of the form

$$\beta_{i,j}(T) = \beta_0 \sqrt{\frac{T_0}{T}} \left( \sum_{k=0}^N a_k T^k \right)$$
(2.32)

where T is the gas temperature,  $T_0$  is a reference temperature and fitting parameters  $a_k$  and values for  $\beta_0$  are given in [76]. The integrated band intensity

$$\alpha_{i,j} = \int_{0}^{\infty} \overline{\kappa}_{\nu} d\nu \tag{2.33}$$

is calculated as function of the temperature from

$$\alpha_{i,j}(T) = \alpha_0 \frac{\left(1 - \exp(-\sum_{k=1}^m u_k \delta_k)\right) \cdot \Psi(T)}{\left(1 - \exp(-\sum_{k=1}^m u_{0,k} \delta_k)\right) \cdot \Psi(T_0)}$$
(2.34)

with parameters for simplified mathematical expressions given in [76] for each band of carbon dioxide and water vapor. In the case of overlapping bands, transmissivities are multiplied

$$\tau_{overlap} = \prod \tau_{i,j}.$$
 (2.35)

Overlapping of bands is found by calculating the band width

$$\Delta\nu_{i,j} = \frac{A_{i,j}}{1 - \tau_{i,j}} \tag{2.36}$$

and looking at lower and upper band limits according to

$$\begin{aligned} \nu_{L,i,j} &= \nu_{C,i,j} - 0.5 \cdot \Delta \nu_{i,j} \\ \nu_{U,i,j} &= \nu_{C,i,j} + 0.5 \cdot \Delta \nu_{i,j} \end{aligned}$$



Figure 2.5: Spectral absorption coefficient of oxy-fuel flue gas with  $\psi_{CO_2} = 0.7$ and  $\psi_{H_2O} = 0.25$  at gas temperature T = 1273.15 K on (a) linear and (b) semi-logarithmic scale according to [76]

or corresponding expressions in case of non–symmetrical bands. The spectral absorption coefficient  $\kappa_{\nu}$  for symmetrical bands can be calculated from

$$\kappa_{\nu} = \frac{\alpha_{i,j}}{\omega_{i,j}} \exp^{-2\left(\frac{|\nu_{C,i,j}-\nu|}{\omega_{i,j}}\right)}.$$
(2.37)

Expressions for asymmetrical bands are given in [76]. Figure 2.5 shows spectral absorption coefficient as function of wavenumber for typical oxy-fuel flue gas composition with  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  ( $\Psi = 5/14 \approx 0.357$ ) at gas temperature T = 1273.15 K on linear and semi-logarithmic scale. Calculation of the spectral absorption coefficient is based on 1 cm<sup>-1</sup> discretization of the wavenumber between 0 and 8000 cm<sup>-1</sup>. The model is described in detail in reference [135] and a stepwise procedure for calculation of emissivity is given in reference [76]. Emissivities for typical carbon dioxide and water vapor mole fractions of air-fired and oxy-fuel systems are presented in section 4.3.1.

#### 2.2.4 Coupling of Emissivity Correlation and RTE

The assumption of gray gas and equality of (spectrally averaged) emission and absorption coefficients<sup>8</sup> lead to the gray RTE formulation [76, 139], equation 2.17.

<sup>&</sup>lt;sup>8</sup>This assumption is only valid if temperatures are constant [137] or only weak temperature gradients occur along the optical path [139].

Gray emissivity model and differential RTE solution method have to be coupled by deriving a spectrally independent absorption coefficient  $\bar{\kappa}$  from modeled gray gas emissivity. According to the rearranged equation 2.23

$$\kappa_{\nu} = -\frac{1}{L}\ln\left(1 - \epsilon_{\nu}\right) \tag{2.38}$$

from section 2.2.2, absorption coefficient  $\kappa_{\nu}$  and emissivity  $\epsilon_{\nu}$  are related to each other so that  $\kappa_{\nu}$  decreases with increasing optical path length<sup>9</sup>.

However, there have been investigations showing that no single gray absorption coefficient  $\overline{\kappa}$  could characterize a non-gray gas as gray [137]. The reason is that one must not spectrally average the RTE itself but directional averages of the spectral RTE [137]. Planck mean absorption coefficient  $\kappa_p$  and Rosseland mean absorption coefficient  $\kappa_r$  are valid only for optically thin (almost transparent) and optically thick (almost black) media [76, 137], respectively.

A gray mean absorption coefficient for variable gas opacity has been proposed by Patch [95],  $\kappa_{pa}$ . However, it is very common to apply an absorption coefficient  $\kappa_e$  based on equation 2.38 which is referred to as *effective absorption coefficient* in the following. Table 2.1 shows the calculation rules of these mean absorption coefficients.

The Planck mean absorption coefficient is based on the Planck black body radiation energy density distribution as weighting function. It is valid only in optically thin (i.e., weakly absorbing) medium and small path lengths [76]. As the characteristic path length becomes greater than Planck mean free path  $L_p = 1/\kappa_p$ , gas reabsorbs radiation emitted from itself [76]. In combustion modeling, the mean geometric path length of radiation is often larger than  $L_p$ . Therefore, Planck mean absorption coefficient is not suitable to calculate heat radiation from large gas volumes [75].

Rosseland mean absorption coefficient [104] is valid in optically thick media [76] (i.e., almost all radiation is reabsorbed). It is based on the temperature derivative of Planck black body radiation distribution as weighting function. Rosseland mean absorption coefficient is applicable where radiative exchange occurs mainly between neighboring volume elements [76].

In engineering calculations, an effective absorption coefficient according to equation 2.38 with gray gas emissivity calculated from either global or spectral emissivity

<sup>&</sup>lt;sup>9</sup>More precisely, *pressure path length* since concentration of radiating gas species in the gas mixture must be considered.

Coefficient	Definition	Validity
Planck mean	$\kappa_p = \frac{\int\limits_0^\infty \kappa_\nu E_{b,\nu} d\nu}{\sigma_c T^4}$	Optical thin limit
Rosseland mean	$\frac{1}{\kappa_{r}} = \frac{\int\limits_{0}^{\infty} \kappa_{\nu}^{-1} \frac{\partial E_{b,\nu}}{\partial T} d\nu}{\int\limits_{0}^{\infty} \frac{\partial E_{b,\nu}}{\partial T} d\nu}$	Optical thick limit
Effective	$\kappa_e = -\frac{1}{L}\ln(1-\epsilon_g)$	Depends on $L$ , $\psi_{CO_2}$ , $\psi_{H_2O}$
Patch	$\kappa_{pa} = \frac{\int\limits_{0}^{\infty} \kappa_{\nu} E_{b,\nu} \cdot e^{-\kappa_{\nu} L} d\nu}{\int\limits_{0}^{\infty} E_{b,\nu} \cdot e^{-\kappa_{\nu} L} d\nu}$	All path lengths

# Table 2.1: Spectrally averaged absorption coefficients for the coupling of emissivity correlation and RTE

models instead of  $\epsilon_{\nu}$  [139] is commonly used and L usually taken as mean optical path length [49] of the regarded system calculated from

$$L = \frac{3.6V}{A} \tag{2.39}$$

where V is the volume and A is the surface area of the gas volume. Good agreement between experimentally obtained heat fluxes and heat fluxes calculated using  $\kappa_e$ have been reported [2]. The latter and also the simple application in the gray formulation of the RTE are major reasons to use the effective absorption coefficient [76]. However, it is more rigorous to use *Patch absorption coefficient* [95] which approaches Planck mean absorption coefficient in the thin limit and decreases with increasing path length. Both, effective and Patch absorption coefficient depend on optical path length [76]. Lallemant and coworkers [75] show that effective absorption coefficient and Patch absorption coefficient are very similar at path lengths  $\leq 0.01$  m for  $\psi_{CO_2} = 0.1$  and  $\psi_{H_2O} = 0.2$  and, thus, substituting  $\kappa_e$  for  $\kappa_{pa}$  does not introduce significant error in the temperature range 1000 to 2000 K. Both coefficients remain in good agreement (factor of two) for path lengths < 1 m but differ significantly at path lengths > 1 m. It will be shown in section 4.3.4 that the agreement between both coefficients depends on carbon dioxide and water vapor mole fractions as well as optical path length. Patch's mean absorption coefficient is expected to be more accurate at intermediate<sup>10</sup> opacity than Planck, Rosseland and effective absorption coefficient [139].

In recent studies [32, 90, 93], the RTE is solved by replacing the spectral absorption coefficient  $\kappa_{\nu}$  by WSGGM absorption coefficients  $\kappa_{u}$  and spectral black body intensity  $I_{b,\nu}(T)$  by  $a_{\epsilon,u}I_{b}$  in the gray formulation of the RTE, equation 2.17:

$$\frac{dI_u}{ds} = \kappa_u (a_{\epsilon,u} I_b - I_u). \tag{2.40}$$

Summation of all  $dI_u/ds$  gives

$$\frac{dI}{ds} = \sum_{u}^{U} \kappa_u (a_{\epsilon,u} I_b - I_u).$$
(2.41)

This procedure where heat transfer is calculated separately for each gray gas and the transparent gas is sometimes referred to as "non-gray" although the assumption of equality of emission and absorption coefficient remains. Becher et al. [8] refer to this method as "banded formulation" of the RTE and state that the approach is not depending on "the correct choice of optical path length". The method can be regarded as a stepwise solution of radiative heat transfer for different optical thicknesses (from transparent to opaque), thus, avoiding to find a single gray absorption coefficient. This approach is not further regarded here since its computational effort is higher and increases with increasing number of gray gases u.

<sup>&</sup>lt;sup>10</sup>Neither optical thin nor optical thick limit.

# **3** Furnace Measurements

For investigation of the influence of oxy-fuel atmospheres on radiative transfer and to validate modeling results, radiative heat fluxes at the walls of an oxy-fuel furnace are measured. Comparing measured and modeled radiative heat fluxes provides information on suitability of the applied emissivity models. Measurements of gas temperature, flue gas species and burn-out behavior serve as boundary conditions. Distribution of temperatures, gas species and particle concentration is derived from measurements and fixed so that the simulation is limited to the calculation of radiative heat transfer. Time-consuming iterative solutions of radiative transfer are not required since the distribution of temperatures is known from measurements. Radiative heat fluxes, total heat fluxes, temperatures and flue gas species are measured in six different cases including air-blown combustion. Ash samples are taken to investigate burn-out behavior during air-firing and oxy-fuel operation.

# 3.1 Test Facility

The regarded test facility is a pre-dried lignite fired oxy-fuel boiler. Heat flux measurements are performed at the boiler front wall. The facility features all major components of oxy-fuel power plants for carbon capture. The facility has been described in several recent publications [13, 68, 101, 129, 130]. Table 3.1 shows the ultimate analysis (as received) of the fuel which is pre-dried lignite from the East German Lausitz area. Volatile content of the fuel is 45.7 % and the share of fixed carbon amounts to 38.5 % (as received). The lower heating value of the fuel is 21.68 MJ/kg.

The test facility is one of the largest plants worldwide to be operated under oxy-fuel conditions. The thermal power input amounts to 30 MW. A single down-shot burner is located at the top of the furnace. The burner throat opening is  $D_0 = 900$  mm in diameter. The dimensions of the furnace are 4.2 m × 4.2 m × 11.8 m. There are six over fire air nozzles located at the side walls at a distance of 7.3 m to the top.

Several measurement openings are located at the front wall of the furnace. It is a natural circulation type boiler. Wall temperatures are independent of furnace height and approximately at evaporation temperature. Compared to commercial boilers, the proportion of thermal input to surface area is relatively low (< 160 kW/m<sup>2</sup>). Figure 3.1 shows a schematic diagram of the boiler and the positions of measurement levels. The flue gas outlet is at the bottom of the rear wall and its dimensions are  $2.8 \times 4.1$  m. Here flue gas enters into the second boiler pass where three convective heating surfaces (superheater 1, superheater 2 and economiser) are installed.

Fuel can be burned with a mixture of oxygen and recirculated flue gas. A cryogenic air separation unit supplies oxygen. Flue gas is recirculated downstream of the electrostatic precipitator and upstream of the drying and desulfurization units (i.e., "wet recirculation"). Thus, water vapor concentration in the flue gas is high compared to recirculation downstream of the flue gas condenser [25]. The boiler can also be operated with air-blown combustion resulting in lower carbon dioxide and water vapor concentration in the flue gas.



Figure 3.1: Simplified scheme of the regarded oxy-fuel test facility

Table 3.2 shows the test cases and the respective oxygen concentration  $y_{O_2}$  of the

С	Н	Ν	S	Ο	Moisture	Ash
$57.5 \ \%$	4.16~%	0.63~%	0.61~%	21.3~%	10.2~%	5.60%

Operation modes Measurement Air Oxy24 Oxy28 Oxy30Oxy32 Oxy36Gas Temperature  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$ Flue gas species ./ √ ./ √ Radiative heat flux ./  $\checkmark$ ./ ./ ./ Total heat flux  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$  $\checkmark$ Burn out sampling  $\checkmark$ √  $y_{O_2} \, [\text{m}^3/\text{m}^3 \, (\text{wet})]$ 0.2070.2420.2790.2990.3180.357Thermal input [MW] 27.226.926.825.926.726.7

 Table 3.1: Ultimate fuel analysis of predried Lausitz lignite (analysis as received)

Table 3.2: Overview on operation modes and furnace measurements

oxidant gas mixture. The table shows which measurements are performed in each of the test cases. In case of combustion in air, there are only total heat flux but no radiative heat flux measurements available due to operational restrictions of the test facility.

# 3.2 Heat Transfer

Heat transfer measurements serve as reference values for comparison with calculated incident heat fluxes (see chapter 5). Comparison of measured and calculated heat flux values provides information on the suitability of applied models used for the calculation of flue gas optical properties. Measurements are performed at different vertical distances from the furnace top. Openings designed for the introduction of probes are located on each furnace level at the center of the boiler front wall.

Two kinds of measurement devices are applied: a hollow ellipsoidal radiometer to measure incident radiative heat flux and a conductivity plug-type heat flux meter to measure total heat flux by radiation and convection. At the tip of the latter probe a certain share of incident radiative heat is reflected according to its surface conditions.



Figure 3.2: Basic principle of (a) hollow ellipsoidal radiometer and (b) conductivity plug-type heat flux meter

The conductivity plug-type meter is applied only in air-blown combustion. Incident radiative heat fluxes are derived from measured total heat fluxes in this case.

# 3.2.1 Radiative Heat Flux

Incident radiative heat flux is measured with the help of a hollow ellipsoidal radiometer designed by IFRF (i.e., the total amount of radiative energy falling on a surface element from a solid angle of  $2\pi$  sr). There is a small circular orifice at the top of the water-cooled probe through which incident radiation enters into an ellipsoidal cavity. Radiation is focussed on a hemispherical receiver pellet by the highly reflective surface of the cavity. Reflectivity of the surface is close to unity due to a thin polished gold coating. The receiver is blackened and absorbs almost all incoming radiation. Constantan wires form a thermocouple giving a voltage U proportional to the energy received by the absorbing pellet. Figure 3.2(a) shows the basic principle of this probe. During measurement, the cavity is constantly purged with nitrogen to prevent deposits and condensation of combustion products on the reflecting surface. A detailed description of this measurement device is given in [18].

# 3.2.2 Total Heat Flux

The design of the conductivity plug-type heat flux meter is described in detail in [18]. With this kind of probe it is possible to measure heat flux by radiation and convection. A cylinder with a receiving surface towards the furnace is located at the top of the probe. The cylinder is surrounded by concentric guard rings with thin

layers of air between them, see figure 3.2(b). This ensures heat conductivity taking place only in axial direction and radial heat exchange being very low. All incident and not reflected radiation on the receiving surface of the cylinder plus the convective heat flux are measured. The receiving surface emits heat radiation according to its temperature. The received heat flux thus comprises following terms:

$$q_t = \epsilon_0 q_{rad} - \epsilon_0 \sigma_c T_0^4 + q_{conv} \tag{3.1}$$

where  $q_t$  is the measured heat flux,  $\epsilon_0$  the emissivity of the receiving surface,  $q_{rad}$  the hemispherical radiative heat flux and  $q_{conv}$  the convective heat flux. Temperature of the receiving surface  $T_0$  is measured by a thermocouple which is installed close to it. There are two more thermocouples installed in order to measure the temperature inside the cylinder at different distances from the receiving surface. Emissivity  $\epsilon_0$  is assumed to be 0.85 according to stainless steel material characteristics [18].

#### 3.2.3 Calibration

The hollow ellipsoidal radiometer does not give absolute values but has to be calibrated with the help of an adiabatic, isothermal black body furnace [18]. Figure 3.3 shows the principle of such a device. The black body furnace is electrically heated. Temperature inside the furnace  $T_b$  is measured with a thermocouple and can be controlled by electric power input. The probe is introduced into the furnace under real measurement conditions (i.e., water-cooled and purged with nitrogen). Radiation from the black body furnace can be calculated from [18]

6

$$q_b = \sigma_c T_b^4. \tag{3.2}$$



Figure 3.3: Black body furnace



Figure 3.4: Calibration measurements of hollow ellipsoidal radiometer and approximation function according to equation 3.3

During calibration measurements, heat flux  $q_b$  is in the range of typical values of incident radiative heat fluxes at the wall of the test facility described in section 3.1. These values are rather low in comparison with commercial boilers (< 160 kW/m<sup>2</sup>). Each value of  $q_b$  corresponds to a certain voltage U. Figure 3.4 shows recorded values from calibration of the hollow ellipsoidal radiometer for furnace temperatures between  $T_b = 200 \text{ °C} (q_b \approx 2.84 \text{ kW/m}^2)$  and  $T_b = 1000 \text{ °C} (q_b \approx 150 \text{ kW/m}^2)$ . Measurement values in the relevant range (33.3 to 112.9 kW/m<sup>2</sup>, see table 3.3 in section 3.2.4) are indicated by larger symbols. The approximative calibration curve derived from measurements in the black body furnace is

$$q_{rad} = 86 \cdot U^{0.69} \tag{3.3}$$

and shown in figure 3.4 as solid line. Coefficient of determination  $R^2$  of the regression curve is 0.9994 and it is 0.9998 in aforesaid relevant range between 33.3 to 112.9 kW/m<sup>2</sup>.

#### 3.2.4 Measured Heat Fluxes

Heat flux measurements are performed along the furnace height at the centerline of the boiler front wall. Radiative heat flux is measured with the help of the hollow ellipsoidal radiometer (see section 3.2.1). Since there are no radiative heat flux

		Radiative neat nux [kw/m]					
Level	$z_d [{ m mm}]$	$Air^*$	Oxy24	Oxy28	Oxy30	Oxy32	Oxy36
0	700	33.3	_	_	29.6	_	_
1	1870	53.0	52.8	61.7	52.4	63.6	73.6
2	2830	53.9	61.5	69.4	70.5	78.4	_
3	4940	68.7	76.8	85.2	98.0	95.8	_
4	6540	63.0	88.5	80.3	89.9	105.6	112.9
5	7940	62.3	78.4	75.2	97.5	97.8	96.4
6	9140	49.5	_	_	84.6	_	_

Radiative heat flux  $[kW/m^2]$ 

\*calculated according to equation 3.4

Table 3.3: Radiative heat fluxes measured at the centerline of the boiler front wall

measurements available for air-blown combustion, radiative heat fluxes are calculated from measured total heat fluxes. In this case, an assumption is made for the convective term:

$$q_{rad} = \frac{q_t - q_{conv} + \epsilon_0 \sigma_c T_0^4}{\epsilon_0}.$$
(3.4)

Emissivity of the receiving surface is assumed to be 0.85. The convective heat flux is then calculated from the total heat flux

$$q_{conv} = \overline{\eta}_{conv} q_t \tag{3.5}$$

where  $\overline{\eta}_{conv}$  is the average share of convective heat transfer among all the other cases determined from the difference between total and radiative heat flux measurement at the respective furnace level. It is assumed that the convective share of the total heat flux in air-blown combustion is within the range of convective share of the other operation modes. This assumption seems reasonable since the other (oxy-fuel) operation modes cover a wide range of flue gas mass flows. Figure 3.5 shows the average convective share at different distances to the furnace top and the standard deviation. The measurement data is fitted by a third degree polynomial. With the help of the convective share calculated from the polynomial function, radiative heat flux can be calculated according to equation 3.4.

Table 3.3 shows measured radiative heat fluxes to the front wall of the furnace at levels 0 to 6. The variable  $z_d$  is the vertical distance of the measurement position from the furnace top. It can be seen from the table that radiative heat flux at the furnace wall is rather low in operation mode *Air*. Regarding oxy-fuel operation, heat transferred to the furnace walls increases with increasing oxygen concentration in the oxidant gas. This observation is consistent with measured gas temperatures



Figure 3.5: Measured average ratio of convective heat flux  $\overline{\eta}_{conv}$  and standard deviation at different distances to the furnace top (×-symbols) and a third degree polynomial data fit (solid line).

presented in the following section. However, there are some exceptions: at some measurement levels, measured heat fluxes of test case Oxy28 are lower compared to case Oxy24. This may be due to operational fluctuations in firing capacity (i.e., fuel supply) during the measurement. For test case Oxy36, there are no measured values available at furnace levels 2, 3 and 6 due to operational reasons.

# 3.3 Other Measurements

Gas temperatures and concentration of radiating gas species are measured in each case presented in table 3.2. The measurements serve as basis for analytical expressions representing temperature and concentration distribution of radiating gas species in section 4.2. Fly ash samples are extracted from the furnace in order to understand burn–out behavior in both air and oxy–fuel combustion.

# 3.3.1 Gas Temperatures

A suction pyrometer is used to measure gas temperatures at different furnace levels and provide information on temperature distribution inside the furnace. For detailed description of this measurement device see reference [18]. In section 4.2.1, analytical

			Gas temperature $[^{\circ}C]$					
Level	$z_d [{ m mm}]$		Air	Oxy30	Oxy24	Oxy28	Oxy32	Oxy36
0	700	max. min.	$878 \\ 650$	$1203 \\ 455$	_	_	_	_
1	1870	max. min.	976 780	$\begin{array}{c} 1203 \\ 634 \end{array}$	$\begin{array}{c} 992\\618 \end{array}$	$\begin{array}{c} 1121 \\ 650 \end{array}$	$\begin{array}{c} 1300 \\ 650 \end{array}$	$\begin{array}{c} 1430 \\ 667 \end{array}$
2	2830	max. min.	$\begin{array}{c} 1106 \\ 829 \end{array}$	$\frac{1268}{764}$	$\begin{array}{c} 1122 \\ 652 \end{array}$	$1285 \\ 667$	$\begin{array}{c} 1341 \\ 715 \end{array}$	$\begin{array}{c} 1414 \\ 699 \end{array}$
3	4940	max. min.	$\begin{array}{c} 1073 \\ 943 \end{array}$	$\begin{array}{c} 1301 \\ 821 \end{array}$	$1268 \\ 797$	$1353 \\ 829$	$1382 \\ 829$	$\begin{array}{c} 1447 \\ 830 \end{array}$
5	7940	max. min.	$\begin{array}{c} 1057 \\ 894 \end{array}$	$1236 \\ 927$	$\begin{array}{c} 1203 \\ 846 \end{array}$	$\begin{array}{c} 1233 \\ 862 \end{array}$	$\begin{array}{c} 1317\\ 860 \end{array}$	$\begin{array}{c} 1382 \\ 890 \end{array}$
6	9140	max. min.	$1073 \\ 780$	$1297 \\ 813$	$\begin{array}{c} 1138\\764\end{array}$	$\begin{array}{c} 1131 \\ 760 \end{array}$	$1220 \\ 759$	$1317 \\ 765$

 Table 3.4: Minimum and maximum gas temperatures measured between front wall and furnace center on furnace levels 0 to 3, 5 and 6 with the probe introduced into the furnace at the center of the front wall

			$\psi_{CO_2}$ [-]		$\psi_{H_2O}$ [-]	
Level	$z_d [{ m mm}]$		Air	Oxy30	Air	Oxy30
0	700	max. min.	$\begin{array}{c} 0.14 \\ 0.08 \end{array}$	$0.62 \\ 0.31$	$\begin{array}{c} 0.11 \\ 0.07 \end{array}$	$\begin{array}{c} 0.28 \\ 0.16 \end{array}$
1	1870	max. min.	$\begin{array}{c} 0.14 \\ 0.08 \end{array}$	$\begin{array}{c} 0.56 \\ 0.50 \end{array}$	$\begin{array}{c} 0.13 \\ 0.07 \end{array}$	$0.29 \\ 0.25$
2	2830	max. min.	$\begin{array}{c} 0.15 \\ 0.11 \end{array}$	$0.59 \\ 0.45$	$\begin{array}{c} 0.13 \\ 0.09 \end{array}$	$0.29 \\ 0.23$
3	4940	max. min.	$0.15 \\ 0.11$	$0.59 \\ 0.45$	$\begin{array}{c} 0.11 \\ 0.07 \end{array}$	$     \begin{array}{c}       0.31 \\       0.23     \end{array} $
4	6540	max. min.	$\begin{array}{c} 0.16 \\ 0.12 \end{array}$	$0.57 \\ 0.52$	$\begin{array}{c} 0.11 \\ 0.09 \end{array}$	$\begin{array}{c} 0.31 \\ 0.28 \end{array}$
5	7940	max. min.	$\begin{array}{c} 0.14 \\ 0.12 \end{array}$	$\begin{array}{c} 0.60 \\ 0.55 \end{array}$	$\begin{array}{c} 0.11 \\ 0.09 \end{array}$	$0.30 \\ 0.23$
6	9140	max. min.	$0.15 \\ 0.11$	$0.62 \\ 0.55$	$0.11 \\ 0.09$	$     \begin{array}{c}       0.31 \\       0.26     \end{array} $

Table 3.5: Minimum and maximum mole fractions of carbon dioxide and water vapor from test cases Air and Oxy30 measured between front wall and furnace center on furnace levels 0 to 6 with the probe introduced into the furnace at the center of the front wall

expressions of temperature distribution are derived from measured values in order to avoid time–consuming iterative calculation of radiative heat transfer. Measured minimum and maximum gas temperatures on furnace levels 0 to 3, 5 and 6 for each operation mode are given in table 3.4.

Lowest maximum temperatures occur in *Air* operation mode. However, minimum temperatures in *Air* are similar to those of oxy–fuel operation or even higher. This indicates different flame shape and lower temperature gradients during air operation compared with oxy–fuel firing. Similar to measured radiative heat flux in the previous section, gas temperatures increase with increasing oxygen concentration in the oxidant gas.

# 3.3.2 Flue Gas Composition

Concentration of carbon dioxide and water vapor in the flue gas is measured in order to find approximate analytical expressions for the concentration distribution of these species. For this purpose, flue gas is extracted at certain positions and the concentration of carbon dioxide and water vapor is measured by IR spectroscopy. Mole fractions of both radiating species are needed as input data for emissivity modeling. Gray solution of the RTE requires homogeneous distribution of radiating gas species along the optical path since spectral distribution of radiation depends on the concentration of participating gas species. However, the measurements show that concentration gradients in the regarded furnace are low except for the near burner region. This is typical for many industrial applications where flame volume occupies only a small part of furnace volume [75]. Gradients of temperature and species concentration are rather small outside the flame volume. Lallemant and coworkers [75] estimate the volume of a 2 MW flame in the IFRF semi-industrial furnace to be 1 m<sup>3</sup> corresponding to approximately 5 % of the furnace volume. Under the assumption that a 30 MW flame fills a volume of  $15 \text{ m}^3$  accordingly, volume share of the flame is approximately 7.2% of the furnace volume. In section 5.4.4, comparisons are made between simulations based on homogeneous and inhomogeneous gas species distribution.

Close to the furnace walls, concentration of carbon dioxide and water vapor is slightly lower and oxygen concentration is higher than at the furnace center. Thus, gas species concentration distribution resembles a cosine function. However, there are regions in the furnace with steep concentration gradients. For example, mole fractions of carbon dioxide and water vapor are much lower in flame regions where intermediate gas species such as carbon monoxide are formed (e.g., near burner region).

Measured minimum and maximum mole fractions of carbon dioxide,  $\psi_{CO_2}$ , and water vapor,  $\psi_{H_2O}$ , are presented in table 3.5. The table shows that both mole fractions are significantly higher for Oxy30 than for Air operation. Mole fractions of carbon dioxide and water vapor are similar for all oxy-fuel operation modes. Carbon dioxide and water vapor concentration in the flue gas is independent of the flue gas recirculation rate except for minor differences caused by different excess oxygen concentrations as the overall stoichiometry is kept constant. Therefore, only measured concentration values of operation mode Oxy30 are shown in table 3.5.

#### 3.3.3 Ash Sampling

Extraction of fly ash samples provides information about the burn-out behavior. Analytical expressions of fuel conversion progress can be derived from these samples. Sampling is done by introducing a probe into the flame (furnace center) and extracting flue gas which is instantly cooled and diluted with nitrogen in order to prevent further combustion progress and condensation of water vapor. The extracted flue gas is then filtered. Particulate matter retained on the filter material is subject to laboratory analysis. The carbon fraction of the ash samples indicates the progress of fuel conversion. Figure 3.6 shows the measured carbon fraction in extracted ash samples at certain distances to the furnace top  $z_d$  in operation modes Air and Oxy30.

Sample analysis shows that combustion in air is characterized by slower fuel conversion than oxy-fuel combustion. This could be caused by higher temperatures in operation case Oxy30 compared to case Air, see table 3.4 in section 3.3.1. Decrease of combustion time with increasing temperatures was found for dry lignite by Kaß and others [62]. Shorter burn-out time in oxy-fuel could also be related to higher partial pressures of carbon dioxide and water vapor. Tappe and Krautz [132] found lower combustion times for pre-dried lignite in  $O_2/CO_2$  atmosphere compared to combustion in air. They related this to the Boudouard reaction (i.e., heterogeneous reaction between carbon and  $CO_2$ ). Al-Makhadmeh and coworkers [3] found significant influence of heterogeneous gasification reactions from pyrolysis experiments in  $CO_2$  atmosphere leading to higher mass release than in N<sub>2</sub> environment.

Measured burn–out behavior is the basis of assumptions made in section 4.2.3 for the distribution of coal and soot particles in the furnace. Carbon in the extracted



Figure 3.6: Measured carbon in ash for cases *Air* and *Oxy30* and exponential functions indicating combustion progress

burn-out samples implies the presence of coal or soot particles at the respective position. Thus, their concentration is related to the progress of carbon conversion which is illustrated in figure 3.6. The figure shows two exponential functions of the form

$$X = X_{max} \cdot e^{K \cdot z_d} \tag{3.6}$$

where X is a variable indicating progress of carbon conversion and, hence, burn–out. Equation 3.6 is applied in section 4.2.3 to model soot and coal particle concentration by substituting X with the respective concentration value and assuming initial coal and soot particle concentration.

Parameter K in equation 3.6 is calculated in the following way:  $X_{max}$  is assumed to be the carbon content in the dry fuel (approx. 63.4 % by weight) representing the initial share of carbon in the fuel before combustion. It is assumed that the fuel's moisture is released and evaporated before ignition of combustible volatiles. With increasing vertical distance to the burner, carbon content in extracted particles decreases. At the end of the furnace, carbon in ash reaches a minimum value  $X_{min}$ which is known from ash samples and approximately 2 % by weight. At this point, the combustion process is incomplete but finished as no further carbon conversion occurs. Parameter K is now determined by minimizing the relative error between measured values of carbon in fuel, ash and samples extracted from the flame on the one hand and the X values computed from equation 3.6 at the respective vertical distances  $z_d$  on the other hand. In order to account for different confidence levels of several coal and ash analyses as opposed to only a small number of burn-out samples<sup>1</sup>, the error between  $X_{max}$  and  $X_{min}$  and their corresponding values calculated from equation 3.6 is weighted by a factor two. Thereby, the fitting curve becomes most precise at  $X_{max}$  and  $X_{min}$ . The resulting exponential functions are shown in figure 3.6. In equation 3.6, K is -0.0004 in case of oxy-fuel combustion and -0.00029 in case of air firing.

The calculated exponential parameters K indicate slightly faster burn-out progress with respect to the vertical distance  $z_d$  under oxy-fuel conditions compared to air firing. This observation is basically in accordance with results on dry lignite oxyfuel combustion [3, 62, 132]. It is assumed in the simulation that K is equal for all oxy-fuel cases despite different gas temperatures.

<sup>&</sup>lt;sup>1</sup>There is a large number of carbon in fuel and carbon in ash analyses available. In contrast, the number of samples extracted from the flame is limited. Therefore, the probability of measured carbon in fuel and carbon in ash to be correct is higher.

# 4 Measurement Based Simulation

The oxy-fuel furnace described in section 3.1 is approximated as rectangular enclosure in a FORTRAN computer program. The enclosure is divided into 208,152 volume elements and 23,352 surface elements resulting in sufficiently small control elements that local thermodynamic properties can be regarded as constant within each. The simulation accounts for radiative properties of carbon dioxide, water vapor and dispersed particles and Monte Carlo ray tracing is used to solve the RTE. This means that a huge number of beams is followed on their way through the enclosure. Gas temperature distribution is fixed according to the measurement results in section 3.3.1. Thus, iterative calculation of radiative transfer is not required since temperatures are known from the beginning. Concentrations of carbon dioxide, water vapor and dispersed particles are also defined in accordance with measurement results. Surface elements are assigned constant temperature of  $235^{\circ}C$  according to the saturated steam temperature of the boiler, thus, neglecting thermal conductivity of tube walls and ash deposits on them for the sake of simplicity. Furnace walls are usually covered by a layer of fly ash deposits so that their optical properties depend mainly on ash optical properties. Reflectivity of surface elements is assumed to be  $\rho_w = 1 - \epsilon_w = 0.15$  in the first place and subject to sensitivity analysis. The calculated incident radiative heat flux is compared with results from heat flux measurements presented in chapter 5.

# 4.1 Monte Carlo Ray Tracing

Ray tracing procedures solve the RTE by following beams on their way through the enclosure. The probability of events like absorption or scattering by gas or particles or reflection at surface elements are described by random numbers according to section 2.1.3. Starting position and direction of each beam are determined by random numbers as well. The procedure is described in detail in the following sections.

#### 4.1.1 Geometry

A rectangular enclosure representing the furnace as highlighted in figure 3.1 is divided into  $42 \times 42 \times 118$  cubic volume elements with edge length d = 100 mm. Within each volume, temperature, gas and particle concentration and absorption coefficient are constant. As a simplification, the furnace is approximated as a rectangular enclosure. Furnace hopper and flue gas outlet are not considered in the simulation. The flue gas outlet is located at the bottom of the furnace wall adjacent to the second boiler pass, see figure 3.1, and its dimensions are  $2.8 \text{ m} \times 4.1 \text{ m}$ =  $11.48 \text{ m}^2$ . The area covers only approximately 5 % of the whole surface area (233.52 m<sup>2</sup>) of the furnace. Moreover, both furnace hopper and flue gas outlet to the second pass are distant from the measurement ports where incident radiation is measured and located in the area of lowest flue gas temperatures so that influence on calculation results is expected to be very limited. Thus, a simple rectangular enclosure representing the furnace geometry appears acceptable. The surface of the enclosure is divided into quadratic surface elements of edge length d = 100 mm. The mean optical path length according to equation 2.39 [49] inside the enclosure is

$$L = \frac{3.6V}{A} = \frac{3.6 \cdot (4.2 \times 4.2 \times 11.8) \text{ m}^3}{233.52 \text{ m}^2} \approx 3.21 \text{ m}$$
(4.1)

Each volume and each surface element is assigned a whole–number index (i, j, k) to define their position in x–, y– and z–direction, respectively, starting in the lower left corner of the enclosure. For example, the volume element with Cartesian coordinates

 $0 \text{ mm} < x_e \le 100 \text{ mm} \quad 0 \text{ mm} < y_e \le 100 \text{ mm} \quad 0 \text{ mm} < z_e \le 100 \text{ mm}$ 

which, if normalized by edge length d, read

$$0 < x_e \le 1$$
  $0 < y_e \le 1$   $0 < z_e \le 1$ 

is assigned an index (i, j, k) = (1, 1, 1). Volume elements are assigned indices  $i \in M_{i,v}$ ,  $j \in M_{j,v}$  and  $k \in M_{k,v}$  with  $M_{i,v} = \{1, \ldots, 42\}$ ,  $M_{j,v} = \{1, \ldots, 42\}$ and  $M_{k,v}[= \{1, \ldots, 118\}$  according to their position in the enclosure. Surface elements are given indices with  $i \in M_{i,s}$ ,  $j \in M_{j,s}$  and  $k \in M_{k,s}$  and  $M_{i,s} = \{0, 43\}$ ,  $M_{j,s} = \{0, 43\}$  and  $M_{k,s} = \{0, 119\}$ . Figure 4.1 shows the subdivision of the enclosure and examples of volume and surface elements. The exemplary volume element in the figure with index (i, j, k) = (22, 27, 52) is located at Cartesian

Level	k [-]	$k^*$ [-]	$z_d \; [mm]$
0	112	7	700
1	100	19	1870
2 3	90 69	29 50	2830 4940
4	53	66	6540
5	39	80	7940
0	27	92	9140

Table 4.1: Measurement levels and corresponding indices k and  $k^*$  of the nearest control elements

coordinates (normalized by edge length d=100 mm)

$$21 < x_e \le 22$$
  $26 < y_e \le 27$   $51 < z_e \le 52$ .

Furnace levels with measurement ports are shown in figure 3.1. Each level corresponds to a certain domain height described by the k-index. Table 4.1 gives an overview on measurement levels, their distance to the furnace top  $z_d$  and respective indices k and  $k^* = 119 - k$ .

#### 4.1.2 Monte Carlo Approach

Radiative heat transfer in the regarded furnace is calculated by Monte Carlo simulation. The Monte Carlo method was developed "as a way of treating problems in which the happenings at a given location are known, at least in the form of statistical distributions, but in which the equations that describe the interactions between locations are extremely difficult to solve" [50]. In the Monte Carlo method applied to radiative transfer, a number of beams is sent through the enclosure containing a certain amount of energy. Location, direction and wavelength of the emitted beam are determined by probability functions representing a physical model which characterizes the radiative process. The energy content of each beam corresponds to local radiative and thermodynamic properties. For a beam emitted from any surface element (i, j, k) it is

$$E = \epsilon(i,j,k)\sigma_c \left(T(i,j,k)\right)^4 \frac{A(i,j,k)}{N(i,j,k)}$$

$$\tag{4.2}$$



Figure 4.1: Subdivision of the furnace with exemplary volume element (22,27,52) and surface element (43,27,99)

where E is the energy content of the beam,  $\epsilon(i, j, k)$  the emissivity, T(i, j, k) the temperature and A(i, j, k) the surface area of the regarded surface element, and N(i, j, k) is the total number of beams which are emitted from surface element (i, j, k). For a beam emitted from any volume element (i, j, k) the energy content is

$$E = 4\kappa(i, j, k)\sigma_c \left(T(i, j, k)\right)^4 \frac{V(i, j, k)}{N(i, j, k)}$$
(4.3)

with  $\kappa(i, j, k)$  being the emission coefficient<sup>1</sup> in the respective control element, V(i, j, k) its volume and N(i, j, k) the total number of beams emitted from it. The number of beams emitted from surface element (i, j, k) can be related to their emissive power by [10, 124]

$$N(i, j, k) = N_s \frac{\epsilon(i, j, k) \left( (T(i, j, k))^4 - T_0^4 \right) A(i, j, k)}{\sum_i \sum_j \sum_k \epsilon(i, j, k) \left( (T(i, j, k))^4 - T_0^4 \right) A(i, j, k)}$$
(4.4)

and similarly for volume elements

$$N(i,j,k) = N_v \frac{\kappa(i,j,k) \left( (T(i,j,k))^4 - T_0^4 \right) V(i,j,k)}{\sum_i \sum_j \sum_k \kappa(i,j,k) \left( (T(i,j,k))^4 - T_0^4 \right) V(i,j,k)}$$
(4.5)

where  $T_0$  is a minimum temperature below which no beams are emitted. Minimum temperature is set close to ambient temperature at 300 K in order to assure that hot control elements emit a large number of beams and cold control elements a smaller number of beams [10]. The number of emitted beams from volume or surface elements depends also on the size of the regarded control element. However, the size of the element can be neglected in equations 4.4 and 4.5 since all volume and all surface elements are of the same size. Hence, the number of beams emitted from a control element depends only on temperature and radiative properties. The total number of emitted beams N is arbitrary but must be large enough to give meaningful results. A number of one billion beams from the volume elements and 10,000 beams from each surface element seems to be sufficient in order to minimize statistical errors and to yield a smooth distribution of radiative heat flux, see section 5.2. The total number of emitted beams from surface and volume elements is  $N_v + N_s =$ N = 1,233,520,000.

<sup>&</sup>lt;sup>1</sup>Equals the absorption coefficient under gray conditions according to equation 2.17.

#### 4.1.3 Ray Tracing

Starting a beam from any place in the enclosure requires definition of its starting coordinates and its direction. The place of emissions if defined by Cartesian coordinates (x, y, z) in x-, y- and z-direction. The point of emission  $P_s(x_s, y_s, z_s)$  of a beam is shown in 4.2. Direction of emission is defined by cone angle  $\Theta$  and circumferential angle  $\Phi$  of the beam. Both angles are calculated on the basis of random numbers. Since a spherical coordinate system is applied in the ray tracing program, cone angle and circumferential angle are converted into spherical coordinates  $\theta$  and  $\phi$  which are polar and azimuthal angle, respectively. Both angles and the spherical coordinate r describing the distance to the fixed origin of the beam are shown in figure 4.2.

The point of emission  $P_s(x_s, y_s, z_s)$  can be calculated with the help of random numbers according to equation 2.18 by

$$x_s = i + R_x - 1$$
  $y_s = j + R_y - 1$   $z_s = k + R_z - 1$ 

with  $R_x$ ,  $R_y$  and  $R_z$  being independent of each other since probability of emission is uniformly distributed in the respective control element due to the essential assumption of constant radiative and thermodynamic properties within each control element. Accordingly,  $R_x$ ,  $R_y$  and  $R_z$  are uniformly distributed in the range 0 < R < 1. At a surface element, two variables define the point of emission. Emission of beams is started consecutively from one control element after another. The number of beams from each of the control elements is defined by equation 4.4 or 4.5. Subsequently, emission direction from either volume or surface element is calculated. Determination of the direction of emission is again based on random numbers. It is



Figure 4.2: Volume element geometry



Figure 4.3: Flow chart of Monte Carlo ray tracing program

shown in [50] that the cone angle of emission from a surface element is

$$\Theta = \arcsin \sqrt{R_{\Theta}} \tag{4.6}$$

under the assumption that the surface emits diffusely. The circumferential angle is

$$\Phi = 2\pi R_{\Phi} \tag{4.7}$$

under the assumption that emission does not depend on  $\Theta$ . Due to the assumption of constant radiative and thermodynamic properties within each control element,  $R_{\Theta}$  and  $R_{\Phi}$  are independent of  $R_x$ ,  $R_y$  and  $R_z$ . From a volume element, beams are emitted into the whole solid angle of  $4\pi$ . Assuming cone and circumferential angle being independent of each other, it is shown in [10] that the cone angle is

$$\Theta = \arccos(1 - 2R_{\Theta}). \tag{4.8}$$

Random numbers are generated and inserted into these equations, step  $\langle i \rangle$  in figure 4.3.

If starting from a wall, cone angle  $\Theta$  and circumferential angle  $\Phi$  are related to the surface normal of the control element from where the beam is emitted or reflected. Both coordinates are therefore converted into spherical coordinates  $\theta$  and  $\phi$  with respect to the applied coordinate system for further calculation. If the point of emission  $P_s$  is located at one of the side walls, the azimuthal angle  $\phi$  is calculated from

$$\phi = \phi_n - \frac{\pi}{2} + \pi R_\Phi \tag{4.9}$$

where  $\phi_n$  is the azimuthal angle of the normal to the surface of the respective wall. The polar angle is then calculated from

$$\theta = 2 \arcsin \sqrt{R_{\Theta}}. \tag{4.10}$$

If  $\phi$  calculated from equation 4.9 is  $\geq 2\pi$  at the wall with  $\phi_n = 2\pi$ ,  $\phi$  is recalculated from

$$\phi = \phi_n - \frac{\pi}{2} + \pi R_{\Phi} - 2\pi. \tag{4.11}$$

Regarding top and ground surface of the furnace, azimuthal angle  $\phi$  is calculated from

$$\phi = 2\pi R_{\Phi} \tag{4.12}$$

and the corresponding polar angle from

$$\theta = \arcsin\sqrt{R_{\Theta}} \tag{4.13}$$

if  $P_s$  is located at the ground surface and

$$\theta = \arcsin\sqrt{R_{\Theta}} + \frac{\pi}{2} \tag{4.14}$$

if  $P_s$  is located at the top of the furnace. If the point of emission is located in one of the volume elements, azimuthal angle  $\phi$  equals  $\Phi = 2\pi R_{\Phi}$  and polar angle  $\theta$  equals  $\Theta = \arccos(1 - 2R_{\Theta})$ .

It is then determined whether the beam is emitted towards positive or negative x–, y– and z– direction and the length  $r_l$  which the beam travels through the *l*-th volume element before entering the next element is calculated, step <ii> in figure 4.3:

$$r_l = \min\left(r_{l,x,max}; r_{l,y,max}; r_{l,z,max}\right)$$

with

$$r_{l,x,max} = \frac{|x_s - i|}{\cos\phi\sin\theta}$$
$$r_{l,y,max} = \frac{|y_s - j|}{\sin\phi\sin\theta}$$
$$r_{l,z,max} = \frac{|z_s - k|}{\cos\theta}$$

where i, j and k are indices of the adjacent volume elements in x–, y– and z–direction, respectively. According to the spherical coordinates  $r, \phi$  and  $\theta$ , the distance to the point where the beam enters the adjacent volume element is calculated from

$$\begin{aligned} x &= r_l \cos \phi \sin \theta \\ y &= r_l \sin \phi \sin \theta \\ z &= r_l \cos \theta. \end{aligned}$$

From point  $P(x_s + x, y_s + y, z_s + z)$ , the regarded beam travels through the next volume element. Figure 4.2 shows a beam entering a control volume and passing through it. The procedure repeats until the beam is absorbed  $\langle iii \rangle$  or scattered  $\langle iv \rangle$  in the gas-particle-suspension or reaches one of the walls  $\langle v \rangle$  where it is either absorbed  $\langle vii \rangle$  or reflected  $\langle vi \rangle$ , see figure 4.3. The traveled distance of

the beam is the sum of distances  $r_l$  through each volume element before absorption, reflection or scattering

$$s = \sum_{l} r_l. \tag{4.15}$$

A beam arriving at a surface element is absorbed if the inequality

$$\epsilon_w \ge R_{\epsilon_w}$$
 (4.16)

is fulfilled. It means that the beam is absorbed if wall emissivity is greater than the generated random number which is uniformly distributed between 0 and 1 representing the probability of absorption. Wall emissivity  $\epsilon_w$  equals absorptivity according to Kirchhoff's law in LTE, equation 2.22, and it is assumed to be 0.85 at first and then subject to sensitivity analysis.

If the gas-particle-suspension absorbs the energy content of the beam prior to its arrival at a surface element, the distance traveled before absorption is calculated again based on a random number in order to determine the volume element where absorption occurs. Howell and Perlmutter [52] show that the distance traveled before absorption can be found by a random number according to

$$R'_{\kappa_g} = 1 - exp\left(-\sum_l \tau_{g,l}\right) \tag{4.17}$$

with the optical thickness  $\tau_{q,l}$  of each incremental element being

$$\tau_{g,l} = r_l \kappa_{g,l} \tag{4.18}$$

where  $r_l$  is the path length which the beam travels through the respective element and  $\kappa_{g,l}$  is the constant absorption coefficient of element l. The summation is carried out over all elements through which the beam travels on its way through the enclosure. The random number  $R'_{\kappa_g}$  in equation 4.17 represents the ratio  $I/I_0$  in Lambert–Beer's law, equation 2.3 in section 2. It is the ratio between intensity Iof the beam after passing through the medium and its intensity  $I_0$  before passing through it. Due to the character of the random number, this ratio is uniformly distributed between 0 (i.e., full absorption) and 1 (i.e., no absorption). The path length  $r_l$  after which the beam is absorbed can then be found from the inequality

$$\sum_{l} r_l \kappa_{g,l} > \ln \frac{1}{R_{\kappa_g}} \tag{4.19}$$
with  $R_{\kappa g} = 1 - R'_{\kappa g}$  and each incremental element having a constant absorption coefficient which is true in LTE. If  $R'_{\kappa g} = 0$ , the right hand side of the inequality becomes zero and absorption occurs after an infinitesimal small distance  $r_l$ . If  $R'_{\kappa g} =$ 1, the right hand side of the equation becomes infinite and  $r_l$  has to be infinite to satisfy the inequality. The beam is tallied at the volume element <viii> where absorption takes place.

Scattering is treated in the same manner based on the scattering coefficient  $\sigma$ 

$$R'_{\sigma} = 1 - exp\left(-\sum_{l} \sigma_{l} r_{l}\right) \tag{4.20}$$

where  $R'_{\sigma}$  now represents the ratio between intensity of the beam I after passing through the medium due to out-scattering and its original intensity  $I_0$ . The beam is scattered if the inequality

$$\sum_{l} \sigma_l r_l > \ln \frac{1}{R_{\sigma}} \tag{4.21}$$

is satisfied with  $R_{\sigma} = 1 - R'_{\sigma}$ . The beam is scattered rather than absorbed if the distance  $r_l$  determined from equation 4.21 is smaller than that calculated from equation 4.19. If the regarded beam is scattered  $\langle iv \rangle$ , a new direction is determined according to equation 4.6 and equation 4.7, step  $\langle i \rangle$  in figure 4.3 (i.e., scattering is assumed to be isotropic).

Surface elements either absorb or reflect  $\langle vi \rangle$  arriving beams according to equation 4.16. A reflected beam is given a new direction  $\langle i \rangle$  based on a random number according to equation 4.6 and equation 4.7 (i.e., surfaces are assumed to reflect diffusely with angle of incidence and reflection being unequal). After reflection, the whole procedure repeats. If a beam is absorbed  $\langle viii \rangle$ , it is tallied at the respective surface element.

Ray tracing continues until N beams have been emitted  $\langle vii \rangle$  from the control elements.

## 4.2 Temperatures and Gas Species Concentrations

The absorption coefficient of each control element must be determined before the ray tracing procedure. Optical properties of each volume element depend on temperature and concentration of radiating gases and particles. These values are derived from furnace measurements in order to avoid computational expensive iterative solutions of the RTE and calculation of flow and chemical reaction processes. The following sections show how required parameters are approximated based on measured values.

#### 4.2.1 Temperature Distribution

Before starting the ray tracing subroutine, the temperature distribution at each level k is approximated by cosine expressions. These expressions are based on measured minimum and maximum gas temperatures on the particular furnace level approximated by polynomial functions of the form

$$T_{g,max}(z) = \sum_{n=0}^{2} \alpha_{T,n} (k^* d - \frac{1}{2} d)^n - 273.15K$$
(4.22)

and

$$T_{g,min}(z) = \sum_{n=0}^{2} \beta_{T,n} (k^* d - \frac{1}{2} d)^n - 273.15K$$
(4.23)

where  $\alpha_{T,n}$  and  $\beta_{T,n}$  are the coefficients listed in table 4.2, d is the edge length of control elements (100 mm), k is the number of the regarded volume element in z-direction and  $k^* = 119 - k$ . Parameters  $\alpha_{T,n}$  and  $\beta_{T,n}$  define quadratic functions which approximate the course of measured minimum and maximum temperatures from furnace top to furnace bottom in a least-square sense and, thus, represent the vertical temperature distribution inside the furnace.

In horizontal direction, cosine–shaped gas temperature distribution is calculated in the intervals  $i_{min} \leq i \leq i_{max}$  and  $j_{min} \leq j \leq j_{max}$  with  $i_{min} = j_{min} = 0$  and  $i_{max} = j_{max} = 42$ . After calculating both maximum and minimum gas temperature from equation 4.22 and equation 4.23, the difference (i.e., the temperature spread) between both values

$$\Delta T_{g,max,min}(k) = T_{g,max}(k) - T_{g,min}(k)$$

and the arithmetic mean of both values

$$\overline{T}_{g,max,min}(k) = \frac{T_{g,max}(k) + T_{g,min}(k)}{2}$$

(a) Horizontal temperatures of test case Oxy30, level 1, k = 100, distance from the furnace top  $z_d = 1870$  mm



(b) Horizontal temperatures of test case Oxy30, level 6, k = 27, distance from the furnace top  $z_d = 9140$  mm



Figure 4.4: Normalized cosine temperature distribution of test case Oxy30 at (a) furnace level 1 ( $k = 100, z_d = 1870$  mm) and (b) furnace level 6 ( $k = 27, z_d = 9140$  mm); normalized temperature 0 equals 0 °C, normalized temperature 1 equals 1626 °C

- (a) Horizontal temperatures of test case (b) Horizontal temperatures of test case Oxy30, level 1, k = 100, distance from the furnace top  $z_d = 1870 \text{ mm}$ 
  - Oxy30, level 3, k = 69, distance from the furnace top  $z_d = 4940 \text{ mm}$



Figure 4.5: Measured (symbols) and modeled (solid line) gas temperatures of test case Oxy30 along furnace width at the furnace center (j = 21) at (a) furnace level 1 ( $k = 100, z_d = 1870$  mm, near burner region) and (b) furnace level 3 ( $k = 69, z_d = 4940 \text{ mm}$ )

(a) Horizontal temperatures of test case (b) Horizontal temperatures of test case Air, level 1, k = 100, distance from the furnace top  $z_d = 1870 \text{ mm}$ 

Air, level 3, k = 69, distance from the furnace top  $z_d = 4940 \text{ mm}$ 



Figure 4.6: Measured (symbols) and modeled (solid line) gas temperatures of test case Air along furnace width at the furnace center (j = 21) at (a) furnace level 1 ( $k = 100, z_d = 1870$  mm, near burner region) and (b) furnace level 3 ( $k = 69, z_d = 4940 \text{ mm}$ )

are determined. Local gas temperature is then calculated from

$$T_g(i,j,k) = \overline{T}_{g,max,min}(k) + \frac{\Delta T_{g,max,min}(k)}{2} \times \cos\left[\left(\frac{1}{2} - \sqrt{\left(\frac{1}{2} - i^*\right)^2}\right) \left(\frac{1}{2} - \sqrt{\left(\frac{1}{2} - j^*\right)^2}\right) 4\pi + \pi\right]$$
(4.24)

where  $i^*$  and  $j^*$  are normalized positions at the center of the respective volume element

$$i^* = \frac{i - i_{min} + 0.5}{i_{max} - i_{min} + 1}$$

and

$$j^* = \frac{j - j_{min} + 0.5}{j_{max} - j_{min} + 1}.$$

At all distances to the furnace top smaller than 1870 mm (118  $\geq k \geq$  100), cosine distribution of temperatures is calculated only in the intervals between  $i_{min} = j_{min} = 15$  and  $i_{max} = j_{max} = 28$ . Outside of these intervals, all control elements are assigned the minimum gas temperature  $T_g(i, j, k) = T_{g,min}(k)$ . This is done in order to account for small temperature gradients in horizontal direction outside the flame and steep gradients in the near burner region between the hot flame and the colder surrounding as revealed by temperature measurement. Temperature gradients in horizontal direction become less steep with increasing distance to the furnace top.

Figure 4.4 shows two exemplary normalized temperature distributions of test case Oxy30 at furnace level 1 and 6. Given values are normalized by maximum<sup>2</sup> gas temperature (1626 °C). Figure 4.5 and figure 4.6 show measured and approximated gas temperatures on furnace level 1 (k = 100, near burner region) and furnace level 3 (k = 69) of test cases Oxy30 and Air, respectively.

There are some deviations as can be seen from figure 4.6(a) where measured values are somewhat higher than calculated temperatures. Increasing the polynomial degree n in the approximative functions 4.22 and 4.23 could further improve the match between measured and calculated values. However, the figures show that the approximated temperature distribution is basically in good agreement with the measured values.

<sup>&</sup>lt;sup>2</sup>This is the maximum temperature resulting from simulation in section 6

		n = 0	n = 1	n = 2
Air		$8.683 \cdot 10^2$	$8.015 \cdot 10^{-2}$	$-6.628 \cdot 10^{-6}$
Oxy30	$\alpha_{T,n}$	$1.188 \cdot 10^{3}$	$2.866 \cdot 10^{-2}$	$-2.134 \cdot 10^{-6}$
Oxy24		$6.901 \cdot 10^2$	$1.964 \cdot 10^{-1}$	$-1.628 \cdot 10^{-5}$
Oxy28		$8.672 \cdot 10^2$	$1.717 \cdot 10^{-1}$	$-1.583 \cdot 10^{-5}$
Oxy32		$1.120 \cdot 10^{3}$	$8.713 \cdot 10^{-2}$	$-7.945 \cdot 10^{-6}$
Oxy36		$1.353 \cdot 10^{3}$	$3.477 \cdot 10^{-2}$	$-3.879 \cdot 10^{-6}$
Air		$5.706 \cdot 10^2$	$7.044 \cdot 10^{-2}$	$-4.817 \cdot 10^{-6}$
Oxy30		$3.691 \cdot 10^2$	$1.399 \cdot 10^{-1}$	$-9.231 \cdot 10^{-5}$
Oxy24	β <sub>T</sub>	$3.823 \cdot 10^2$	$1.327 \cdot 10^{-1}$	$-9.775 \cdot 10^{-5}$
Oxy28	PI, n	$3.780 \cdot 10^2$	$1.300 \cdot 10^{-1}$	$-3.251 \cdot 10^{-6}$
Oxy32		$5.087 \cdot 10^2$	$6.776 \cdot 10^{-2}$	$-1.003 \cdot 10^{-5}$
Oxy36		$5.579 \cdot 10^2$	$4.981 \cdot 10^{-2}$	$-3.334 \cdot 10^{-6}$

**Table 4.2:** Coefficients  $\alpha_{T,n}$  and  $\beta_{T,n}$  of the gas temperature polynomials  $T_{g,max}(z)$  and  $T_{g,min}(z)$  for each of the modeled test cases

		n = 0	n = 1	n = 2
Air Oxy30	$\alpha_{CO_2,n}$	$\frac{1.408 \cdot 10^{-1}}{6.231 \cdot 10^{-1}}$	$\frac{1.247 \cdot 10^{-6}}{-2.223 \cdot 10^{-5}}$	$0 \\ 2.415 \cdot 10^{-9}$
Air Oxy30	$\beta_{CO_2,n}$	$\frac{8.133 \cdot 10^{-2}}{3.445 \cdot 10^{-1}}$	$4.930 \cdot 10^{-6} 4.362 \cdot 10^{-5}$	$0 -2.395 \cdot 10^{-9}$
Air Oxy30	$\alpha_{H_2O,n}$	$\frac{1.223 \cdot 10^{-1}}{2.799 \cdot 10^{-1}}$	$-1.485 \cdot 10^{-6}$ 7.268 \cdot 10^{-6}	0 -4.519 $\cdot 10^{-10}$
Air Oxy30	$\beta_{H_2O,n}$	$7.128 \cdot 10^{-2} \\ 1.722 \cdot 10^{-1}$	$2.602 \cdot 10^{-6} \\ 2.582 \cdot 10^{-5}$	$0 - 1.921 \cdot 10^{-9}$

**Table 4.3:** Coefficients  $\alpha_{g,n}$  and  $\beta_{g,n}$  of the concentration polynomials  $\psi_{g,max}(z)$ and  $\psi_{g,min}(z)$  for carbon dioxide and water vapor

### 4.2.2 Gas Species Concentration

Concentration of radiating gaseous species is calculated in a similar manner as gas temperature. Measured maximum and minimum concentrations at the center of a control element are approximated by equations of the form

$$\psi_{g,max}(k) = \sum_{n=0}^{2} \alpha_{g,n} (k^* d - \frac{1}{2} d)^n$$
(4.25)



Figure 4.7: Measured (symbols) and modeled (solid line) mole fractions, test case Oxy30, on furnace level 3 (k = 69, distance from furnace top 4940 mm): (a) carbon dioxide,  $\psi_{CO_2}$ , and (b) water vapor,  $\psi_{H_2O}$ , along furnace width

and

$$\psi_{g,min}(k) = \sum_{n=0}^{2} \beta_{g,n} (k^* d - \frac{1}{2} d)^n$$
(4.26)

where g stands for either CO<sub>2</sub> or H<sub>2</sub>O. Table 4.3 shows coefficients applied in test cases *Air* and *Oxy30*. The coefficients of the latter case are applied in each oxy-fuel test case since measurements show that gas species concentration is very similar among them. The concentration at a volume element with index (i, j, k) is then determined from

$$\psi_g(i,j,k) = \overline{\psi}_{g,max,min}(k) + \frac{\Delta\psi_{g,max,min}(k)}{2} \times \cos\left[\left(\frac{1}{2} - \sqrt{\left(\frac{1}{2} - i^*\right)^2}\right) \left(\frac{1}{2} - \sqrt{\left(\frac{1}{2} - j^*\right)^2}\right) 4\pi + \pi\right]$$
(4.27)

with

$$\overline{\psi}_{g,max,min}(k) = \frac{\psi_{g,max}(k) + \psi_{g,min}(k)}{2}$$

and

$$\Delta \psi_{g,max,min}(k) = \psi_{g,max}(k) - \psi_{g,min}(k).$$

Figures 4.7(a) and (b) show that the cosine functions sufficiently represent measured concentrations of carbon dioxide and water vapor. Strong gradients, however, may be not accurately approximated particularly in the near burner region. The error (a) Horizontal distribution of carbon dioxide concentration, test case Oxy30, level 0, k = 112, distance from the furnace top  $z_d = 700$  mm



(b) Horizontal distribution of carbon dioxide concentration, test case Oxy30, level 6, k = 27, distance from the furnace top  $z_d = 9140$  mm



Figure 4.8: Normalized cosine carbon dioxide concentration profiles of test case Oxy30 at furnace level 0 (k = 112) and furnace level 6 (k = 27); normalized mole fraction 0 equals 0, normalized mole fraction 1 equals 0.62

introduced by using equation 4.27 is expected to be small since strong concentration gradients are located where coal and soot particles dominate radiation.

Figure 4.8 shows concentration profiles of carbon dioxide at furnace level 0 (k = 112) and level 6 (k = 27) normalized by maximum<sup>3</sup> mole fraction 0.62. It becomes clear from these figures that the distribution of carbon dioxide concentration becomes more uniform with higher distances from the burner (i.e., smaller values of k).

The ratio between water vapor and carbon dioxide mole fraction is calculated from

$$\Psi(i,j,k) = \frac{\psi_{H_2O}(i,j,k)}{\psi_{CO_2}(i,j,k)}$$

The mole fraction ratio  $\Psi$  serves as input parameter for the applied global emissivity model.

### 4.2.3 Radiative Properties of Dispersed Particles

Coal and char, fly ash and soot particles are the most important solid particles dispersed in the gas phase of pulverized coal combustors. They emit, absorb and scatter heat radiation [66, 92]. In order to achieve reasonable simulation results, it is mandatory to consider the optical properties of dispersed particles in the calculation of radiative heat transfer. It is assumed that scattering of radiation by the particle phase is isotropic and that coal particles are monodispersed which means that all coal particles have the same average diameter. Polydispersed ash particles are represented by two fractions of particles with either small (fly ash) or large (residual ash) particle size. Spectrally averaged absorption coefficients for coal and ash particles are then calculated as well as a mean absorption coefficient for soot particles according to Felske and Tien [35].

In order to calculate the radiative properties of solid particles, their complex index of refraction  $z_C$  must be known [92]. The complex index of refraction comprises the refractive index,  $x_C$  and the absorptive index  $y_C$  so that  $z_C = x_C - y_C i$  where *i* is the imaginary unit.

Optical properties of soot have been investigated by many researchers [24, 42, 81, 92, 123]. A mean absorption coefficient can be calculated by choosing appropriate

<sup>&</sup>lt;sup>3</sup>This maximum carbon dioxide mole fraction results from simulation in section 6.

spectral average values of  $x_C$  and  $y_C$ . The spectral absorption coefficient of soot can be written as [17]

$$\kappa_{\lambda} = \frac{36\pi}{\lambda} f_{v,s} \frac{x_C y_C}{(x_C^2 - y_C^2 + 2)^2 + 4x_C^2 y_C^2} = \frac{C_0 f_{v,s}}{\lambda}$$
(4.28)

where  $f_{v,s}$  is the soot volume fraction (to be determined later), and  $C_0$  is a constant depending on the complex index of refraction. Chang and Charalampopoulos [16] presented polynomial expressions for  $x_C$  and  $y_C$  in the wavelength range  $0.4 \cdot 10^{-6} \text{ m} \leq \lambda \leq 30 \cdot 10^{-6} \text{ m}$ :

$$x_C(\lambda) = 1.811 + 0.1263 \ln \lambda + 0.0270 \ln^2 \lambda + 0.0417 \ln^3 \lambda$$
(4.29)

$$y_C(\lambda) = 0.5821 + 0.1213 \ln \lambda + 0.2309 \ln^2 \lambda + 0.0100 \ln^3 \lambda.$$
 (4.30)

Figure 4.9 shows the refractive index  $x_C$  and the absorptive index  $y_C$  as functions of wavelength. The spectrum is divided into wavelength blocks of  $\Delta \lambda = 0.1 \cdot 10^{-6}$  m and mean values of  $x_C$  and  $y_C$  are then determined from

$$\overline{x_C} = \sum_{\lambda=0.4}^{30} x_C(\lambda) \left[ F(\lambda + \frac{1}{2}\Delta\lambda) - F(\lambda - \frac{1}{2}\Delta\lambda) \right]$$
(4.31)

$$\overline{y_C} = \sum_{\lambda=0.4}^{30} y_C(\lambda) \left[ F(\lambda + \frac{1}{2}\Delta\lambda) - F(\lambda - \frac{1}{2}\Delta\lambda) \right]$$
(4.32)

where  $\lambda$  is the center of a wavelength block and  $F(\lambda)$  is the fractional function of black body radiation. An approximation of  $F(\lambda)$  is taken from the literature [16] as

$$F(\lambda) = \frac{15}{\pi^4} \sum_{n=1}^{4} \left[ \frac{e^{-n\chi}}{n} \left( \chi^3 + \frac{3\chi^2}{n} + \frac{6\chi}{n^2} + \frac{6}{n^3} \right) \right]$$
(4.33)

with  $\chi = C_2/\lambda T_{\chi}$  for  $\chi \geq 2$ . Temperature  $T_{\chi}$  is assumed to be 1400 K which is the rounded average gas temperature  $T_g$  of volume elements where soot particles are present  $(f_{v,s} > 0)$ . Temperature of soot  $T_s$  and gas temperature  $T_g$  are usually equal due to small particle sizes [92] so that it is justified to use the average gas temperature as  $T_{\chi}$ . For values of  $\chi < 2$ , the fractional function of black body radiation is approximated by

$$F(\lambda) = 1 - \frac{15}{\pi^4} \chi^3 \left( \frac{1}{3} - \frac{\chi}{8} + \frac{\chi^2}{60} - \frac{\chi^4}{5040} + \frac{\chi^6}{272160} - \frac{\chi^8}{13305600} \right)$$
(4.34)

according to [118] resulting in a spectrally averaged complex refractive index  $\overline{z_C}$  =

2.0998 - 1.1061i. An average, gray absorption coefficient is calculated according to Felske and Tien [35]

$$\kappa_s = \frac{3.72 f_v \overline{C_0} T_s}{C_2} \tag{4.35}$$

where  $\overline{C_0}$  is

$$\overline{C_0} = \frac{36\pi \overline{x_C y_C}}{\left(\overline{x_C}^2 - \overline{y_C}^2 + 2\right)^2 + 4\overline{x_C}^2 \overline{y_C}^2} = 3.1589$$

$$(4.36)$$

and  $C_2 = 1.4387770 \cdot 10^{-2}$  mK, the second Planck constant. Under the assumption of soot volume fraction  $f_{v,s}$  being  $10^{-7}$  (which is in the typical range [74]) and  $T_{\chi} = 1400$  K, the average soot absorption coefficient  $\kappa_s$  becomes 0.1982 m<sup>-1</sup>. It is 0.1975 m<sup>-1</sup> if  $T_{\chi} = 1000$  K. Apparently, influence of temperature  $T_{\chi}$  in the fractional function of black body radiation on soot absorption coefficient is rather small so that the assumption of  $T_{\chi}$  being the average gas temperature in volume elements with  $f_{v,s} > 0$  seems to be acceptable.

Soot volume fraction is calculated in a similar manner as gas temperature within  $15 \leq i \leq 28$  and  $15 \leq j \leq 28$  from cosine functions of  $i^*$  and  $j^*$ . The function describing the soot volume fraction  $f_{v,s}(i, j, k)$  is

$$f_{v,s}(i,j,k) = \overline{f}_{v,s,max,min}(k) + \frac{1}{2}\Delta f_{v,s,max,min}(k) \times \left[ \left(\frac{1}{2} - \sqrt{\left(\frac{1}{2} - i^*\right)^2}\right) \left(\frac{1}{2} - \sqrt{\left(\frac{1}{2} - j^*\right)^2}\right) 4\pi + \pi \right]$$
(4.37)

with

$$\overline{f}_{v,s,max,min}(k) = \frac{f_{v,s,max}(k) + f_{v,s,min}(k)}{2}$$

and

$$\Delta f_{v,s,max,min}(k) = f_{v,s,max}(k) - f_{v,s,min}(k).$$

Maximum soot concentration  $f_{v,s,max}(k)$  is derived from a function of the form

$$f_{v,s,max}(k) = f_{v,s,0} \cdot e^{K(k^*d - 0.5d)}$$
(4.38)

which corresponds to the approximation function of carbon conversion progress in section 3.3.3 with K being either -0.0004 for oxy-fuel or -0.00029 for air-blown combustion, respectively. Carbon content in burn-out samples indicates how much combustible fuel substance remains at the respective distance to the burner. Soot formation in turn happens only in fuel-rich environments as a result of incomplete



Figure 4.9: The refractive index  $x_C$  and the absorptive index  $y_C$  of soot as a function of wavelength according to [16]

combustion [92].

The initial soot volume fraction  $f_{v,s,0}$  in equation 4.38 is assumed to be  $10^{-6}$  since typical values are between  $10^{-9}$  and  $10^{-6}$  [74]. Section 5.4.2 presents a variation of  $f_{v,s,0}$  within this range showing that this assumption has only minor influence on heat flux results. The minimum soot volume fraction is assigned a constant value  $f_{v,s,min}(k) = 0$ .

Scattering by soot particles is neglected in the simulation. This is justified since particle size of soot is small with respect to the wavelengths in the infrared [92].

Volume fraction of coal particles  $f_{v,c}(i, j, k)$  is calculated similar to soot volume fraction, equation 4.37, as a function of  $i^*$  and  $j^*$  in the intervals  $15 \le i \le 28$  and  $15 \le j \le 28$ . The maximum coal concentration  $f_{v,c,max}(k)$  is calculated similar to equation 4.38:

$$f_{v,c,max}(k) = f_{v,c,0} \cdot e^{K(k^*d - 0.5d)}$$
(4.39)

with K according to the functions given in section 3.3.3. The initial coal volume fraction  $f_{v,c,0}$  is assumed to be

$$f_{v,c,0}(k) = \frac{\dot{m}_c}{\rho_c \cdot \dot{v}_g} \tag{4.40}$$

where  $\dot{m}_c$  is the coal mass flow to the burner,  $\rho_c$  the coal density (1300 kg/m<sup>3</sup>)

Mesh size $d_c$ [m]	Residual coal mass $m_{c,r}$ [%]
$500 \cdot 10^{-6}$	0.2
$200 \cdot 10^{-6}$	6.4
$90 \cdot 10^{-6}$	30.0
$40 \cdot 10^{-6}$	58.0
$20 \cdot 10^{-6}$	78.5

Table 4.4: Particle size distribution

and  $\dot{v}_g$  is the volume flow of oxidant gas. The value of  $f_{v,c,min}(k)$  is zero. Values for complex index of refraction of lignite are presented in [140]. Approximative relations for spectrally independent absorption and extinction coefficients are given in [9] based on data of Buckius and Hwang [11] for averaged particle radii. As a simplification, coal particles are assumed to be spherical and monodisperse. Different average diameters of monodisperse coal particles can be calculated. Sauter diameter  $d_{c,s}$  represents the average sphere size which corresponds to the specific surface of all particles [127]. The particle size distribution is presented in table 4.4 where  $d_c$ is the mesh size of the analysis sieve and  $m_{c,r}$  the share of coal remaining on the sieve.

According to the measured particle size distribution, six particle size classes l are defined (0 to 20, 20 to 40, 40 to 90, 90 to 200, 200 to  $500 \cdot 10^{-6}$  m and 0.5 to 2 mm). The upper limit of the class with highest particle sizes is chosen arbitrarily. However, the share of particles belonging to this class is very small and particles with diameter greater than 2 mm are very rare. The mass of coal particles belonging to each class  $m_{c,l}$  is determined according to the size distribution in table 4.4. The average diameter  $d_{c,l}$  of each class l is then calculated (10, 30, 65, 145, 350 \cdot 10^{-6} m and 1.25 mm, respectively) and weighted by the corresponding fuel mass fraction  $m_{c,l}/m_c$ 

$$d_{c,m} = \sum_{l=1}^{6} d_{c,l} \frac{m_{c,l}}{m_c}.$$
(4.41)

giving the mean particle diameter [127],  $d_{c,m} = 82.5 \cdot 10^{-6}$  m, which is used as average diameter for an equivalent monodisperse particle phase in the simulation. Since radiative transfer is related to the surface of coal particles, it is also possible to use the square root of mean square diameter [127] of each class l

$$\overline{d_c^2} = \sum_{l=1}^{6} \left( d_{c,l} \right)^2 \frac{m_{c,l}}{m_c}.$$
(4.42)

as average diameter which is  $118.3 \cdot 10^{-6}$  m according to the particle size distribution given in table 4.4. Sauter diameter is calculated from [127]

$$d_{c,s} = \frac{\overline{d_c^3}}{\overline{d_c^2}} \cdot \frac{1}{f_h} \tag{4.43}$$

with

$$\overline{d_c^3} = \sum_{l=1}^6 \left( d_{c,l} \right)^3 \frac{m_{c,l}}{m_c} \tag{4.44}$$

and  $f_h$  being the *Heywood factor* which characterizes the relation between the actual specific surface of a particle and the specific surface of an equivalent sphere (e.g., with the same volume). Heywood factor of pulverized coal is 1.8 to 2.1 [43]. If  $f_h = 2$ , Sauter diameter becomes  $121.3 \cdot 10^{-6}$  m according to the particle size distribution shown in table 4.4 and is higher than  $d_{c,m}$ , see table 4.5.

Heat flux results of test case Oxy30 are shown in section 5.4.2 based on mean particle size  $d_{c,m}$  and Sauter diameter  $d_{c,s}$ . It can be seen from the figure that the influence on wall heat fluxes is rather low so that application of mean particle diameter  $d_{c,m}$ in the simulation seems acceptable.

According to [9], the absorption coefficient of monodispersed coal particles is calculated from

$$\kappa_c = f_{A,c} \left[ \left( 0.0032 \left( 1 + \left( \frac{\Phi_c}{425} \right)^{1.8} \right) \right)^{-\frac{6}{5}} + \left( \frac{10.99}{\Phi_c^{0.02}} \right)^{-\frac{6}{5}} \right]^{-\frac{5}{6}}$$
(4.45)

and the extinction coefficient from

$$\beta_c = f_{A,c} \left[ \left( 0.0032 \left( 1 + \left( \frac{\Phi_c}{650} \right)^{2.0} \right) \right)^{-\frac{5}{4}} + \left( \frac{13.75}{\Phi_c^{0.13}} \right)^{-\frac{5}{4}} \right]^{-\frac{4}{5}}.$$
 (4.46)

The total projected area of the particle cloud  $f_{A,c}$  appearing in the equations is

$$\frac{d_{c,m} \text{ [m]}}{82.5 \cdot 10^{-6}} \frac{\sqrt{\overline{d_c^2}} \text{ [m]}}{118.3 \cdot 10^{-6}} \frac{d_{c,s} \text{ [m]}}{121.3 \cdot 10^{-6}}$$

Table 4.5: Average particle diameters

calculated from

$$f_{A,c} = \frac{\pi}{4} \frac{\overline{d_c}^2 \cdot \dot{m}_c}{\rho_c \cdot \dot{v}_g} \left( \frac{4}{3} \pi \left( \frac{\overline{d_c}}{2} \right)^2 \right)^{-1}$$
(4.47)

where  $\Phi_c$  is the size parameter defined as

$$\Phi_c = \frac{3f_{v,c}}{4f_{A,c}}T_c \tag{4.48}$$

with  $T_c$  being the temperature of coal particles. Since there is no information available on particle temperature, it is assumed in the modeling that coal particles and gas phase have the same temperature. The influence of particle temperature on  $\kappa_c$ is approximately linear: increase of particle temperature  $T_c = 1273$  K by factor 1.1 ( $T_c = 1400$  K) results in an increase of absorption coefficient by factor 1.11. Decrease of temperature by factor 0.9 ( $T_c = 1146$  K) results in a reduction of absorption coefficient by factor 0.89. Factor between measured particle and gas temperatures in a pulverized coal combustor reported in reference [100] are between 0.84 and 0.99 and on average 0.95. The only exception is a measurement value taken directly at the fuel inlet. Measured particle temperature is here 25 % lower than gas temperature.

Absorption and extinction coefficients of ash particles are calculated according to equations 4.45 and 4.46, respectively. Polydisperse fly ash particles are approximated as two monodisperse particle phases. These are referred to as residual ash and fly ash. According to [47, 66, 109], each char particle forms three to five coarse ash particles with 10 to  $30 \cdot 10^{-6}$  m diameter and 200 to 500 fine ash particles in diameter range 1 to  $10 \cdot 10^{-6}$  m. However, mass of three spherical ash particles of diameter  $20 \cdot 10^{-6}$  m ( $3.77 \cdot 10^{-12}$  g) or  $30 \cdot 10^{-6}$  m ( $12.72 \cdot 10^{-12}$  g) is higher than the mass of ash in a spherical coal particle ( $2.14 \cdot 10^{-12}$  g) with diameter  $d_{c,m} = 82.5 \cdot 10^{-6}$  m (density of ash  $\rho_a = 3000 \text{ kg/m}^3$ ). Therefore, it is assumed that residual ash has particle diameter  $d_{ra} = 10 \cdot 10^{-6}$  m (mass of four particles is  $0.63 \cdot 10^{-12}$  g). Fly ash is assumed to have an average diameter  $d_{fa} = 1.5 \cdot 10^{-6}$  m representing a particle size distribution that is consistent with the assumption on residual ash diameter and the mass of ash in a spherical coal particle coal particle with  $d_{c,m} = 82.5 \cdot 10^{-6}$  m.

Assuming ash density of  $\rho_a = 3000 \text{ kg/m}^3$ , total projected area of residual ash  $f_{A,ra}$ and fly ash  $f_{A,fa}$ , their volume fractions  $f_{v,ra}$  and  $f_{v,fa}$ , and their size parameters  $\Phi_{ra}$  and  $\Phi_{fa}$  can be calculated. Volume fraction of both ash particle phases is assumed to be equally distributed in the whole furnace. Ash particle temperature is assumed to be the same as gas temperature in each control element.

# 4.3 Gas Emissivity

Gas emissivity  $\epsilon_g$  is calculated as the intensive quantity describing the ability of the gas mixture in each control element to emit and absorb heat radiation according to Kirchhoff's law in LTE. This is done by either a weighted sum of three gray gases and one clear gas (WSGG3+1) model with coefficients according to [122] or a weighted sum of four gray gases and one clear gas (WSGG4+1) model with coefficients represented by functions of the mole fraction ratio

$$\Psi(i,j,k) = \frac{\psi_{H_2O}(i,j,k)}{\psi_{CO_2}(i,j,k)}$$
(4.49)

according to the approach presented in reference [56].

The EWBM serves as benchmark model for the curve fitting procedure producing suitable WSGG4+1 coefficients. Comparisons are made between WSGG4+1 and other recently published emissivity correlations.

However, solving the RTE requires an extensive quantity describing gas absorption as function of path length. In case of gray solutions, a mean absorption coefficient is determined for that purpose based on the applied WSGG correlation. It is shown in the following how various approaches yield different results of the absorption coefficient. The absorption coefficient is based on average optical path length according to the geometry of the furnace, see section 4.1.3, instead of a mean optical path length according to the geometry of control elements. The latter method depends on the chosen spatial discretization which is a known disadvantage [32].

### 4.3.1 EWBM Emissivities

Calculating emissivity  $\epsilon_g$  from the EWBM is based on dividing the spectrum into a number of K = 8000 spectral blocks with block width  $\Delta \nu_k = 1 \text{ cm}^{-1}$ . Each spectral block is assigned a transmissivity  $\tau_k$  which is calculated according to the band transmissivity at the regarded block or according to equation 2.35 if bands overlap. Emissivity is then calculated from [76]:

$$\epsilon_g = \sum_{k=1}^{K} (1 - \tau_k) \left( F(\nu_{L,k}, T) - F(\nu_{U,k}, T) \right)$$
(4.50)



Figure 4.10: EWBM band transmissivities at their spectral location at T = 1273.15 K and L = 3.21 m for (a) water vapor and (b) carbon dioxide regarding different mole fractions  $\psi_{H_2O}$  and  $\psi_{CO_2}$ 

where  $\nu_{L,k}$  and  $\nu_{U,k}$  are the lower and upper wavenumber limits of the block, respectively, and  $F(\nu, T)$  is the fractional black body function, equations 2.11 and 2.12.

Figure 4.10 shows band transmissivities  $\tau_{i,j}$  of water vapor and carbon dioxide at their spectral location under the assumption of T = 1273.15 K, L = 3.21 m and different mole fractions. It can be seen from the figure that gas transmissivity decreases with increasing mole fraction of radiating species. The highest decrease in transmissivity from air-blown to oxy-fuel combustion results for the 960 and  $1060 \text{ cm}^{-1}$  bands of carbon dioxide. Transmissivity of both bands is 0.9 if the mole fraction  $\psi_{CO_2}$  is 0.15 which is a typical concentration of carbon dioxide in the flue gas of air-fired systems. Increased mole fraction of carbon dioxide  $\psi_{CO_2} = 0.7$  as in oxy-fuel systems leads to transmissivities of 0.539 and 0.453, respectively.

Figure 4.11 shows EWBM spectral emissivity  $\epsilon_{\nu}$  as function of wavenumber  $\nu$  based on path length L = 3.21 m and gas temperature T = 1273.15 K. Figure 4.11(a) shows spectral emissivities of a gas mixture with  $\psi_{CO_2} = 0.15$  and  $\psi_{H_2O} = 0.1$ which can be considered as typical flue gas mixture of air-fired combustion. Figure 4.11(b) shows spectral emissivities if  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  as an example of flue gas from oxy-fuel combustion. Both figures include the fractional black body function  $F(\nu, T)$  according to equations 2.11 and 2.12. The figures show that spectral emissivities increase with increasing mole fraction of radiating species. There are less transparent regions at elevated mole fractions of carbon dioxide and water vapor. Hence, emissivity of the gas mixture calculated according to equation 4.50 increases as can be seen from figure 4.12(a). This figure shows emissivity  $\epsilon_g$  of both gas mixtures as function of optical path length. It can be seen from figure 4.12(b) that the deviation between emissivity of both gas mixtures is highest at path lengths between 0.0001 and 0.001 m. However, relative deviation between emissivities of both gas mixtures remains between 35 and 15 % for relevant path lengths 1 m  $< L \leq 10$  m. At a mean path length of L = 3.21 m (which is the mean optical path length of the regarded test furnace), modeled emissivity of oxy-fuel flue gas is approximately 30 % higher than that of air-firing based on aforesaid carbon dioxide and water vapor concentrations.

### 4.3.2 WSGGM Coefficients

WSGG3+1 coefficients presented by Smith and coworkers [122] are shown in table 4.6. They are often used in engineering software tools to calculate flue gas emissivity. The correlation was found to be in best agreement with EWBM generated emissivity among several models [76]. Comparison in reference [76], however, is limited to maximum molar fractions of 0.2 for both, carbon dioxide and water vapor. In the flue gas of oxy-fuel boilers, both molar fractions possibly exceed this value leading to deviations in terms of emissivity in comparison with benchmark models such as the EWBM. Figure 4.13 and figure 4.14 show emissivities generated from Smith's WSGG3+1 correlation and from EWBM as function of path length at 1000 and 1500 K gas temperature, respectively. It is shown in figure 4.13(a) and 4.14(a) that Smith's emissivity correlation is in very good agreement if  $\psi_{CO_2} = 0.15$  and  $\psi_{H_2O} = 0.1$  which corresponds to typical flue gas composition of air-fired systems. From figures 4.13(b) and 4.14(b) it becomes clear that higher mole fractions of carbon dioxide and water vapor lead to significant deviation between global and band model emissivities.

It	is	important	to	remember	that	$_{\mathrm{the}}$	WSGG3+1	l model	considers	only	the	$\operatorname{sum}$	of
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u	$\kappa_u$	$b_{\epsilon,u,1} \cdot 10^1$	$b_{\epsilon,u,2} \cdot 10^4$	$b_{\epsilon,u,3} \cdot 10^7$	$b_{\epsilon,u,4} \cdot 10^{11}$
1	0.4303	5.150	-2.303	0.9779	-1.494
2	7.055	0.7749	3.399	-2.297	3.770
3	178.1	1.907	-1.824	0.5608	-0.5122

**Table 4.6:** Absorption coefficients  $\kappa_u$  and polynomial coefficients  $b_{\epsilon,u,v}$  for WSGG3+1 as provided by Smith et al. [122] for  $\psi_{CO_2} = \psi_{H_2O} = 0.1$  $(\Psi = 1)$ 



Figure 4.11: Spectral emissivities  $\epsilon_{\nu}$  generated from EWBM for L = 3.21 m, T = 1273.15 K and (a)  $\psi_{CO_2} = 0.15$ ,  $\psi_{H_2O} = 0.1$  and (b)  $\psi_{CO_2} = 0.7$ ,  $\psi_{H_2O} = 0.25$  and black body fractional function  $F(\nu, T)$  ("BBF") according to equations 2.11 and 2.12



Figure 4.12: Comparison of emissivities generated from EWBM for typical flue gas of air firing with  $\psi_{CO_2} = 0.15$  and  $\psi_{H_2O} = 0.1$  and oxy-fuel combustion  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  (T = 1273.15 K) as function of path length: (a) absolute values of emissivity and (b) relative deviation of typical air firing emissivity as compared to typical oxy-fuel emissivity on semi-logarithmic scale



(a) Combustion in air at 1000 K:

(b) Oxy-fuel combustion at 1000 K:



Figure 4.13: Emissivity of  $CO_2/H_2O$  gas mixtures as function of path length at 1000 K temperature for (a) combustion in air with  $\psi_{CO_2} = 0.15$  and  $\psi_{H_2O} = 0.1$  and (b) oxy-fuel combustion with  $\psi_{CO_2} + \psi_{H_2O} = 0.95$  and three different ratios  $\Psi$ . Emissivities are generated from EWBM and Smith's [122] emissivity correlation (WSGG3+1). Molar fractions of carbon dioxide and water vapor, respectively, are given in parentheses.





Figure 4.14: Emissivity of  $CO_2/H_2O$  gas mixture as function of path length at 1500 K temperature for (a) combustion in air with  $\psi_{CO_2} = 0.15$  and  $\psi_{H_2O} = 0.1$  and (b) oxy-fuel combustion with  $\psi_{CO_2} + \psi_{H_2O} = 0.95$  and three different ratios  $\Psi$ . Emissivities are generated from the EWBM and Smith's [122] emissivity correlation (WSGG3+1). The molar fractions of carbon dioxide and water vapor, respectively, are given in parentheses.

mole fractions of radiating species. The actual share of water vapor or carbon dioxide is not considered. Deviation between EWBM and WSGG3+1 generated emissivities becomes especially low at high mole fractions of water vapor. This is of particular importance for oxy-fuel combustion systems burning fuels with high water content (e.g., lignite) or high hydrogen content (e.g., natural gas) or in case of wet flue gas recirculation prior to flue gas condensation.

In order to allow for higher molar fractions of carbon dioxide and water vapor, the WSGG4+1 can be applied to calculate the total gas emissivity based on the equation

$$\epsilon_g = \sum_{u=0}^{U} a_{\epsilon,u}(T) \left[ 1 - e^{-\kappa_u pL} \right]$$
(4.51)

with U = 4. Weighting factors  $a_{\epsilon,u}(T)$  are represented each by a temperature polynomial

$$a_{\epsilon,u}(T) = \sum_{v=1}^{V} b_{\epsilon,u,v} T^{v-1}$$
(4.52)

and the weighting factor of the clear gas is defined as

$$a_{\epsilon,0} = 1 - \sum_{u=1}^{U} a_{\epsilon,u}(T).$$
(4.53)

It is demonstrated by Khare and others [65] that an additional gray gas term is required in case of large scale oxy-fuel furnaces. They extended the parameter set provided by Smith et al. [122] with additional coefficients for equation 4.51. These model parameters, however, do not account for actual carbon dioxide and water vapor mole fractions so that several sets of coefficients are required for different flue gas compositions.

Johansson et al. [56] recently published coefficients of a new four gray and one clear gas approach where both, weighting factors and absorption coefficients are linear or polynomial functions of the mole fraction ratio  $\Psi(i, j, k)$ . According to their approach, weighting factors of the WSGG4+1 correlation are represented by polynomial functions

$$a_{\epsilon,u}(T,\Psi) = \sum_{v=1}^{V} \sum_{w=1}^{W} \left( c_{\epsilon,u,v,w} \ \Psi(i,j,k)^{w-1} \right) \ T^{v-1}.$$
(4.54)

with W = 4. The coefficients  $b_{\epsilon,u,v}$  of WSGG4+1 temperature polynomials are

now represented by polynomial functions of the mole fraction ratio which are valid within certain  $\Psi$ -intervals. The polynomial functions form a cubic spline (i.e., a non-uniform, smooth, piecewise polynomial function). WSGG4+1 coefficients are given in appendix A.1.

Piecewise–polynomial functions of mole fraction ratio  $\Psi$  represent the absorption coefficients  $\kappa_u$ :

$$\kappa_u(\Psi) = \sum_{p=0}^{P} d_{u,p} \Psi^{p-1}$$
(4.55)

with P = 4. The coefficients  $d_{u,p}$  are given in appendix A.2. Figure 4.15 illustrates the polynomials  $\kappa_u(\Psi)$  and  $a_{\epsilon,u}(T = 1000 \ K, \Psi)$  of each of the gray gas terms in equation 4.51.

The polynomial coefficients  $c_{\epsilon,u,v,w}$  are determined from a curve fitting procedure minimizing an error function  $f(\Psi, L, T)$  in a least square sense by the Nelder-Mead method [94] with the mole fraction ratio being  $5/90 \le \Psi \le 65/30$ 

$$f(\Psi, L, T) = \sum_{n}^{N} \sum_{m}^{M} \left( \epsilon_{\text{WSGG}}(L_n, T_m, \Psi) - \epsilon_{\text{EWBM}}(L_n, T_m, \Psi) \right)^2$$
(4.56)

where  $\epsilon_{WSGG}(L_n, T_m, \Psi)$  is the WSGG4+1 emissivity at a certain temperature, path length and mole fraction ratio resulting in an objective function

$$f(\Psi, L, T) = \sum_{n}^{N} \sum_{m}^{M} \left( \sum_{u=0}^{U} \sum_{v=1}^{V} \sum_{w=1}^{W} \left[ (c_{\epsilon,u,v,w} \Psi(i,j,k)^{w-1}) \times T^{v-1} \left[ 1 - e^{-pL \sum_{p=0}^{P} d_{u,p} \Psi^{p-1}} \right] \right] - \epsilon_{\text{EWBM}}(L_n, T_m, \Psi) \right)^2$$
(4.57)

with  $\epsilon_{\text{EWBM}}(L_n, T_m, \Psi)$  being the corresponding EWBM emissivity data, cf. section 4.3.1. The curve fit is based on temperatures and path lengths in accordance with reference [122]. The sum of carbon dioxide and water vapor mole fraction is assumed to be  $\psi_{CO_2} + \psi_{H_2O} = 0.95$ , the residual 5 % being non-radiative species as nitrogen

and oxygen. The curve fitting procedure is based on following input data:

$$L [m] = \{0.005, 0.015, 0.05, 0.15, 0.5, 1.5, 5, 15, 50\}$$
  
$$T [K] = \{600, 650, 700, \dots, 2350, 2400\}$$
  
$$\Psi = \{5/90, 6/89, 7/88, \dots, 64/31, 65/30\}.$$

Figure B.1 in appendix B shows emissivities at  $\Psi = 65/30$  and  $\Psi = 5/90$  generated from EWBM and WSGG4+1. Figure B.2 in appendix B shows the same EWBM data and emissivities generated from WSGG3+1 approach using Smith's coefficients. WSGG4+1 emissivities are in better agreement with EWBM data than WSGG3+1 values. Latter emissivities do not increase with increasing path lengths if *L* becomes greater than 15 m. Thus, WSGG3+1 emissivity at 15 m path length is the same as at 50 m. At  $L \approx 5$  m and  $\Psi = 5/90$ , emissivity is clearly overestimated by this correlation.

Discontinuous EWBM data in figure B.1 and figure B.2 (e.g., at T = 2000 K and L = 0.15 m) stems from the application of two different approximative black body distribution functions depending on wavelength as recommended in [76, 118] giving different values at the boundary value  $\chi = C_2 \nu/T = 2$  as described at the beginning of chapter 2.

Table 4.7 contains values of the error function  $f(\Psi, L, T)$ , equation 4.56. It can be seen from the table that values of  $f(\Psi, L, T)$  are significantly lower if the WSGG4+1 correlation is applied instead of WSGG3+1. Accuracy of WSGG4+1 emissivity model is also higher if the mole fraction of non-radiative species is > 5 % (e.g.,  $f(\Psi = 30/55, L, T)$  and  $f(\Psi = 10/75, L, T)$ ). However, accuracy with respect to the EWBM may decrease if the assumption  $\psi_{CO_2} + \psi_{H_2O} = 0.95$  is not fulfilled. Mole fractions of other species such as carbon monoxide and oxygen are especially high in the near burner regions where particle concentration is high and also emission and absorption by particles is dominant. The error introduced by high oxygen or carbon monoxide concentrations in this area is therefore expected to be of minor importance.

Figure 4.16 compares emissivities of oxy-fuel flue gas at T = 1273.15 K with  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  generated from WSGG3+1 and WSGG4+1. Deviation of WSGG3+1 emissivity from WSGG4+1 emissivity remains less than +10 % at relevant path lengths between 0.1 and 10 m. Deviation between both models is slightly higher at higher gas temperature, see figure 4.17.



Figure 4.15: (a) Absorption coefficient  $\kappa_u$  and (b) weighting factor  $a_{\epsilon,u}$  at T = 1000 K as function of mole fraction ratio  $\Psi$  for each of the four gray gases from WSGG4+1 emissivity correlation



Figure 4.16: At T = 1273.15 K: Comparison of emissivity at  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  modeled with WSGG3+1 [122] and WSGG4+1 correlation: (a) emissivity as function of path length and (b) relative deviation of WSGG3+1 emissivity as compared to WSGG4+1 emissivity against the optical path length on semi–logarithmic scale



Figure 4.17: At T = 1573.15 K: Comparison of emissivity at  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  modeled with WSGG3+1 [122] and WSGG4+1 correlation: (a) emissivity as function of path length and (b) relative deviation of WSGG3+1 emissivity as compared to WSGG4+1 emissivity against the optical path length on semi–logarithmic scale

	$\Psi$	65/30	5/90	30/55	10/75
WSGG3+1 WSGG4+1	$\begin{array}{c} f^{3+1}(\Psi,L,T) \\ f^{4+1}(\Psi,L,T) \\ f^{4+1}/f^{3+1} \end{array}$	$\begin{array}{c} 1.50 \cdot 10^{0} \\ 1.92 \cdot 10^{-2} \\ 1.28 \% \end{array}$	$\begin{array}{c} 2.64 \cdot 10^{0} \\ 1.81 \cdot 10^{-2} \\ 0.68 \ \% \end{array}$	$\begin{array}{c} 4.26 \cdot 10^{-1} \\ 1.30 \cdot 10^{-2} \\ 3.05 \% \end{array}$	$\begin{array}{c} 2.64 \cdot 10^{0} \\ 1.91 \cdot 10^{-2} \\ 4.48 \ \% \end{array}$

**Table 4.7:** Values of the error function  $f(\Psi, L, T)$  at different mole fraction ratios  $\Psi$  for Smith's emissivity correlation (WSGG3+1) [122] and the four gray gases plus one clear gas model (WSGG4+1)

### 4.3.3 WSGG4+1 Validation

There are existing gray emissivity relations allowing for different partial pressures of carbon dioxide and water vapor. The model of Leckner [79] is considered "as the most general" [76] gray emissivity model since it can be applied for arbitrary total pressures and partial pressures of carbon dioxide and water vapor [26]. Leiser [82] investigated and recommended Leckner's emissivity correlation for modeling oxyfuel atmospheres. Emissivity is calculated as sum of both species' emissivity and an overlap correction:

$$\epsilon_g = \epsilon_{CO_2} + \epsilon_{H_2O} - \Delta\epsilon \tag{4.58}$$

where the correction term  $\Delta \epsilon$  is a function of path length and partial pressures of carbon dioxide and water vapor. For a detailed description of the model see references [77, 79, 82].

As a result of its validity over a wide range of total pressures and partial pressures of radiating species, Leckner's model appears applicable in case of oxy-fuel combustion as well. Leiser showed in his work [82] that the calculated radiation intensity profiles based on Leckner's model and a wide band correlated-k method [128] are similar in a 100 kW oxy-fuel test furnace [5] with mean path length of approximately 0.64 m. However, the model is limited to partial pressure path lengths of  $p_{CO_2}L < 0.1 \cdot 10^6$  mPa and  $p_{H_2O}L < 1 \cdot 10^6$  mPa [76]. Assuming a total pressure of 0.1 MPa and  $\psi_{CO_2} = 0.07$  (partial pressure of carbon dioxide  $p_{CO_2} = 0.07$  MPa), the mean path length L of the regarded system should not exceed 1.42 m according to these limitations. However, mean path length in the regarded furnace is L = 3.21 m and can be even higher in large-scale coal-fired boilers. Figure 4.18(a) shows the emissivity of a gas mixture at T = 1273.15 K with  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  calculated with the help of Leckner's model as function of optical path length. It can be seen from the figure that the gas emissivity remains constant at path lengths L > 10 m. Compared with WSGG4+1 emissivity correlation, the model underesting the set of the test of the test of the test of the test.

0.1



(a) T = 1273.15 K,  $\psi_{CO_2} = 0.7$ ,  $\psi_{H_2O} = 0.25$ 

Figure 4.18: Comparison of emissivities generated from WSGG4+1 model of section 4.3.2, Yin's model [143], Johansson's models [56, 57], Kangwanpongpan's model [61] and Leckner's model [79]: (a) emissivity as function of optical path length and (b) relative deviation from WSGG4+1 emissivity as function of optical path length on semi-logarithmic scale

Path length, L [m]

10

100

1

timates the gas emissivity due to its limitation to partial pressure path lengths of carbon dioxide  $p_{CO_2}L < 0.1 \cdot 10^6$  mPa. Figure 4.18(b) shows the relative deviation between gas emissivities based on Leckner's model and on WSGG4+1 correlation as function of path length on semi–logarithmic scale. It can be seen that deviation between both models increases at path lengths L > 10 m.

Recently, new WSGG parameters have been published by different authors which take into account partial pressures of carbon dioxide and water vapor of oxy-fuel systems [90]. Johansson and coworkers developed a WSGG model with four gray gases and one clear gas. Weighting factors and absorption coefficients are represented by linear [56] or polynomial [57] functions of water vapor to carbon dioxide mole fraction ratio  $\Psi$ . By this, they allow for different flue gas compositions. They found single linear or polynomial functions for each parameter covering the whole range of  $0.125 \leq \Psi \leq 2$ . The WSGG4+1 model of section 4.3.2 follows this approach calculating the weights and the absorption coefficients from spline polynomials. Yin and coworkers [143] give new parameters for four gray gases and one clear gas at fixed water vapor to carbon dioxide ratios  $\Psi$  (0.125, 0.25, 0.75, 1, 2, and 4). In their model, the sum of partial pressures of carbon dioxide and water vapor equals 1013.3 hPa. The model is valid at optical path lengths  $0.001 \le L \le 60$  m. Kangwanpongpan et al. [61] most recently published coefficients for polynomial functions of molar ratio  $\Psi$  representing absorption coefficients and polynomial coefficients of temperature functions which in turn represent WSGGM weighting factors. Their data fit is based on line-by-line calculations from HITEMP 2010 [106] database instead of EWBM data.

Figure 4.18 compares the WSGG4+1 model of section 4.3.2 and the emissivity correlations described above. The models of Yin [143] and Johansson [56, 57] are in good agreement with the WSGG4+1 correlation. The relative deviation between emissivities is lower than 15 % for relevant path lengths 1 m < L < 10 m. Emissivities calculated from the model of Kangwanpongpan et al. [61] are higher than other results at path lengths > 5 m. This becomes clear also from their publication [61] where comparison is made between their model and others [57, 143]. Their model is based on line-by-line calculations. However, at L = 3.21 m which is the mean path length inside the regarded test furnace, deviation between WSGG4+1 and Kangwanpongpan's model is rather small. Becher et al. [8] made a comprehensive comparison of emissivity results generated from recently developed WSGGM [57, 64, 72, 122, 143] with emissivities calculated from HITEMP 2010 [106] and found the model of Johansson et al. [57] to give most accurate results with maximum deviation of 21 % at high temperatures. All in all, plenty of development work has been done on accurate emissivity modeling with respect to elevated concentrations of carbon dioxide and water vapor and different mole fraction ratios of both species. The WSGG4+1 model yields similar results in comparison with other models which justifies its application in this work.

#### 4.3.4 Mean Absorption Coefficient

EWBM, WSGGM and other correlations have to be coupled with the differential gray RTE formulation by a spectrally averaged absorption coefficient which is an extensive quantity (see section 2.2.4). WSGGM weighting factors  $a_{\epsilon,u}(T, \Psi)$  can be interpreted as sum of blackbody radiation over a group of wavelength bands with identical absorption coefficient  $\kappa_u$  which in turn can be regarded as mean absorption coefficient of these bands [49, 75]. According to this physical interpretation of WSGGM parameters, weights and absorption coefficients are used for an ad hoc calculation of mean absorption coefficients. Planck mean absorption coefficient is then determined from

$$\kappa_p = \frac{\sum_{u=0}^{U} \kappa_u(\Psi) a_{\epsilon,u}(T, \Psi) \sigma_c T^4}{\sigma_c T^4}$$
(4.59)

with a weighted sum of U gray gases. Rosseland mean absorption coefficient can be calculated in the same manner from

$$\frac{1}{\kappa_r} = \frac{\sum_{u=0}^{U} 4\frac{1}{\kappa_u(\Psi)} a_{\epsilon,u}(T,\Psi) \sigma_c T^3}{4 \sigma_c T^3}.$$
(4.60)

It is shown in [75, 76] that Patch absorption coefficient [95] is related to the effective absorption coefficient by

$$\kappa_{pa} = \kappa_e + L \cdot \frac{\partial \kappa_e}{\partial L} \tag{4.61}$$

which simplifies the calculation of Patch absorption coefficient as it can be derived from the effective absorption coefficient. Inserting a sum of gray gases into the equation to calculate  $\kappa_e$ , see table 2.1 in section 2.2.4, gives the effective absorption coefficient

$$\kappa_e = -\frac{1}{L} \ln \left[ 1 - \left( \sum_{u=0}^U a_{\epsilon,u}(T, \Psi) \left( 1 - e^{-\kappa_u(\Psi)pL} \right) \right) \right]$$
(4.62)

and  $\partial \kappa_e / \partial L$  reads

$$-\frac{1}{L^{2}}\left[\frac{pL\sum_{i=0}^{I}-a_{\epsilon,u}\kappa_{u}\cdot e^{-\kappa_{u}pL}}{1-\left(\sum_{u=0}^{U}a_{\epsilon,u}(1-e^{-\kappa_{u}pL})\right)}-\ln\left[1-\left(\sum_{u=0}^{U}a_{\epsilon,u}(1-e^{-\kappa_{u}pL})\right)\right]\right]$$

Regarding the physical interpretation of WSGGM weights  $a_{\epsilon,u}(T, \Psi)$  and absorption coefficients  $\kappa_u(\Psi)$  mentioned above, Patch mean absorption coefficient is calculated from

$$\kappa_{pa} = \frac{\sum_{u=0}^{U} \kappa_u(\Psi) a_{\epsilon,u}(T, \Psi) \sigma_c T^4 \cdot e^{-\kappa_u(\Psi)pL}}{\sum_{u=0}^{U} \sigma_c T^4 \cdot e^{-\kappa_u(\Psi)pL}}.$$
(4.63)

Equation 4.63 gives the same result as equation 4.61 if inserted equation 4.62 and  $\partial \kappa_e / \partial L$ .

Figure 4.19 compares different mean absorption coefficients of a gas mixture at T = 1273.15 K with  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  as function of optical path length on linear scale, figure 4.19(a), and logarithmic scale, figure 4.19(b). Emissivity is calculated from WSGG4+1, see section 4.3.2. It can be seen from the figure that effective and Patch absorption coefficient approach Planck mean absorption coefficient in optically thin media (i.e., only at very small pressure path lengths). This indicates that Planck mean absorption coefficient is not suitable for coupling RTE solution and emissivity model since the mean optical path L in the regarded furnace is 3.21 m according to equation 2.39 in section 2.2.4. Planck mean free path length  $L_p = 1/\kappa_p$  of the regarded gas mixture is 0.123 m ( $\kappa_p = 8.11$  m<sup>-1</sup>) and, thus, < L. This means that the medium can not be regarded as optically thin. Planck mean absorption coefficient is not applicable in case of large volumes such as the modeled furnace since it does not account for radiation emitted and reabsorbed in the gas [75].

Rosseland absorption coefficient is valid only in case of strongly absorbing media and not applicable for the calculation of radiative heat transfer in flames [76]. It is shown in figure 4.19 that Rosseland absorption coefficient is higher than Patch absorption coefficient at path lengths greater than 0.5 m. This is due to the characteristics of Patch absorption coefficient approaching Planck mean absorption coefficient in the thin limit and a minimum absorption coefficient in the thick optical limit [95].



(a) Semi–logarithmic scale:



Path length, L [m]



Figure 4.20: Relative deviation of effective absorption coefficient  $\kappa_e$  from Patch absorption coefficient  $\kappa_{pa}$  against optical path length for a gas mixture at T = 1273.15 K and oxy-fuel atmosphere with  $\psi_{CO_2} = 0.7$ and  $\psi_{H_2O} = 0.25$  ( $\Psi \approx 0.357$ ) using WSGG4+1 and WSGG3+1 emissivity correlations (solid line "Oxy WSGG4+1" and dotted line "Oxy WSGG3+1") and for typical flue gas composition of air firing with  $\psi_{CO_2} + \psi_{H_2O} = 0.25$  using WSGG3+1 emissivity correlation (dashed line "Air WSGG3+1")

Deviation of the effective absorption coefficient  $\kappa_e$  from Patch absorption coefficient  $(\kappa_e - \kappa_{pa})/\kappa_{pa}$  increases with increasing path length as can be seen from figure 4.20, curve "Oxy WSGG4+1" (T = 1273.15 K,  $\psi_{CO_2} = 0.7$ ,  $\psi_{H_2O} = 0.25$ ). The relative deviation of the effective absorption coefficient from Patch absorption coefficient remains within 100 % at path lengths  $\leq 0.1$  m and 200 % at path lengths  $\leq 1$  m.

Patch absorption coefficient is only  $0.26 \cdot \kappa_e$  at the characteristic path length of the regarded furnace L = 3.21 m and the assumptions made for the gas mixture  $(T = 1273.15 \text{ K}, \psi_{CO_2} = 0.7, \psi_{H_2O} = 0.25)$ . This shows that calculation of spectrally averaged absorption coefficients might be more influential on accuracy of wall heat flux predictions than the emissivity correlation. Comparison with figure 4.16 in section 4.3.2 shows that the deviation between WSGG3+1 and WSGG4+1 emissivity correlation remains < 10% at all relevant path lengths. Influence of the applied emissivity correlation is low in comparison with the deviation introduced by using either effective or Patch absorption coefficient.

Several researchers have been working on finding theoretically sound spectrally av-

eraged absorption coefficients for the (semi-)gray formulation of the RTE, equations 2.16 and 2.17. Reference [76] provides a comprehensive overview on this work. Although there is no gray absorption coefficient which could "make a non-grey gas grey" [137], Patch's absorption coefficient yields intensity results which are in the range of -25 to +28 % of spectral calculations in a large number of test cases representing the entire range of optical thickness while Planck mean absorption coefficient yields results between -43 and +455 % [76]. It is shown in [76] that absorption coefficients for intermediate optical thickness have to depend on optical path length L in order to account for the gas' reabsorption of its own emitted radiation. It is expected that Patch absorption coefficient yields more accurate results than the effective absorption coefficient [139] due to its rigorous derivation and since it "is based on a sound theoretical treatment of the RTE" [76]. By contrast, it is not clear whether the effective absorption coefficient "leads to accurate total radiative fluxes predictions" [76].

Figure 4.20 shows the deviation between effective and Patch absorption coefficient for typical air firing flue gas with  $\psi_{CO_2} + \psi_{H_2O} = 0.25$  at T = 1273.15 K. The emissivity is determined by WSGG3+1 and Smith's coefficients [122], curve "Air WSGG3+1". It can be seen that both absorption coefficients are similar at path lengths  $L \leq 0.01$  m. At path lengths below 15 m, deviation of effective absorption coefficient from Patch absorption coefficient remains below 225 %. This includes characteristic path lengths of most commercial boilers according to equation 2.39. In fact, good agreement between measured and modeled wall heat fluxes has been reported in the literature if the effective absorption coefficient is applied in air firing [75]. However, at path lengths L > 15 m, agreement between both coefficients becomes rather poor.

Finally, figure 4.20 presents the deviation of effective from Patch absorption coefficient if emissivity of oxy-fuel flue gas with  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  is modeled with the help of Smith's WSGG3+1 correlation, curve "Oxy WSGG3+1". It can be seen that the deviation between both coefficients increases with increasing path length. In particular at path lengths > 5 m, effective absorption coefficient  $\kappa_e$  becomes significantly (more than 1000 %) higher than  $\kappa_{pa}$ . Hence, combination of WSGG3+1 emissivity modeling and  $\kappa_e$  as extensive quantity is not suitable for radiative heat flux predictions in oxy-fuel boilers with L > 5 m. At L = 3.21 m (mean path length of the regarded test facility), deviation between WSGG3+1 based Patch and effective absorption coefficient is of the same order of magnitude as if based on WSGG4+1 emissivity ( $\kappa_{pa} \approx 0.26 \cdot \kappa_e$ ).



Figure 4.21: Absolute values of derivative of  $\kappa_e$  with respect to L multiplied by L and divided by  $\kappa_e$  (absolute value of relative slope of  $\kappa_e$ ) as a function of optical path length L on semi–logarithmic scale

Figure 4.22(a) compares WSGG3+1 and WSGG4+1 based Patch absorption coefficients as function of optical path length L according to the assumptions made earlier for typical oxy-fuel flue gas. Patch gas absorption coefficient based on Smith's WSGG3+1 model ( $\kappa_{pa}^{3+1}$ ) is significantly lower at path lengths  $L \geq 10$  m. At the characteristic optical path length of L = 3.21 m according to the geometry of the regarded test facility, WSGG3+1 based Patch absorption coefficient,  $\kappa_{pa}^{3+1}$ , is approximately 30 % higher than WSGG4+1 based Patch absorption coefficient,  $\kappa_{pa}^{4+1}$ , as can be seen from figure 4.22(c).

The effective absorption coefficient based on WSGG3+1 correlation,  $\kappa_e^{3+1}$ , and the effective absorption coefficient derived from WSGG4+1 emissivity model,  $\kappa_{pa}^{4+1}$ , are in good agreement at relevant path lengths. Figure 4.22(b) compares both absorption coefficients. Comparison between figure 4.22(a) and 4.22(b) shows that WSGG3+1 and WSGG4+1 based effective absorption coefficients become higher than corresponding Patch absorption coefficients at path lengths L > 1 m. This basically corresponds with the findings presented in reference [75]. Figure 4.22(d) shows the relative deviation between  $\kappa_e^{3+1}$  and  $\kappa_e^{4+1}$  which is rather low even at large optical path lengths  $L \geq 10$  m.

It becomes clear from figure 4.22 that deviation between WSGG3+1 and WSGG4+1 based Patch absorption coefficients is higher than the deviation between WSGG3+1

and WSGG4+1 based effective absorption coefficients. Deviation between  $\kappa_{pa}^{3+1}$  and  $\kappa_{pa}^{4+1}$  becomes very large at optical path lengths  $L \geq 10$  m.

Regarding figures 4.20 and 4.22(c) and (d), the 'oscillating' character of  $\kappa_{pa}$  as function of L is striking. According to the equation  $\kappa_e + L \cdot \partial \kappa_e / \partial L$ , Patch absorption coefficient depends on the derivative of  $\kappa_e$  with respect to L which is not constant. Figure 4.21 shows the absolute value of  $\partial \kappa_e / \partial L$  if multiplied by L and related to  $\kappa_e$ (for better visualization) as function of L on semi–logarithmic scale. The resulting relative slope function of  $\kappa_e$  is not monotonic<sup>4</sup> and reveals the 'oscillating' character of  $\partial \kappa_e / \partial L$  which seems to be an inherent characteristic of the WSGGM.

<sup>&</sup>lt;sup>4</sup>However,  $\partial \kappa_e / \partial L$  is a monotonic decreasing function of L.


Figure 4.22: Comparison between WSGG3+1 and WSGG4+1 based (a) Patch absorption coefficients  $\kappa_{pa}$  and (b) effective absorption coefficient  $\kappa_e$  as functions of path length L for a gas mixture at T = 1273.15 K with mole fractions  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  on double logarithmic scale and respective deviation between WSGG3+1 and WSGG4+1 based absorption coefficients as function of path length L on semilogarithmic scale

# 5 Results

Radiative transfer in the test furnace presented in section 3.1 is calculated as described in chapter 4. The Monte Carlo simulation results show the influence of  $CO_2/H_2O$  rich flue gas on emissivity, average gas absorption coefficient and radiative wall heat fluxes. The effect of different boundary conditions such as the total number of beams, flue gas composition, gas temperature and wall emissivity on radiative heat flux is evaluated. Comparing measured radiative heat fluxes with calculated ones provides information on the performance of applied models and boundary conditions.

## 5.1 Absorption Coefficients

In this section, absorption coefficients according to the flue gas composition of operation modes Air and Oxy30 (see section 4.2) are discussed. In order to limit the investigation to the influence of flue gas composition, other relevant parameters are assumed to be equal in both cases: gas temperature is calculated according to test case Air and particle concentration is taken from test case Oxy30. There is no specific reason to choose particle and temperature distribution in this manner other than to enable comparison of radiative heat fluxes for different flue gas mixtures and to allow a closer view on the influence of carbon dioxide and water vapor concentration on the gas absorption coefficient.

#### 5.1.1 Gas and Particle Absorption Coefficients

Gas emissivity increases with increasing mole fraction of radiating gas species. Therefore, flue gas emissivity of oxy-fuel combustion is higher than that of air-fired furnaces. Increase in gas emissivity leads to higher gas absorption coefficients. Gas absorption coefficient in test case Air is modeled on the basis of WSGG3+1



Figure 5.1: (a) Gas absorption coefficient  $\kappa_g$  and (b) ratio of gas absorption coefficient and particle absorption coefficient  $\kappa_g/\kappa_{part}$  against furnace width (in x-direction) from 0 to 4200 mm ( $1 \le i \le 42$ ) at the furnace center (distance to the wall in y-direction 2100 mm; j = 21) on furnace level 2 (distance to furnace top 2830 mm; k = 90), see also figure 4.1





Figure 5.2: (a) Gas emissivities calculated from WSGG3+1 and WSGG4+1 emissivity correlations as functions of mole fractions of radiative species if  $\Psi = 1$  ( $\psi_{CO_2} = \psi_{H_2O}$ ), T = 1273.15 K, and L = 3.21 m; (b) total gas emissivities calculated from WSGG3+1 model (with  $\psi_{CO_2} = 0.15$  and  $\psi_{H_2O} = 0.1$ ) and WSGG4+1 (with  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$ ) as function of normalized gas temperature (normalized by 1626 °C) at L = 3.21 m



Figure 5.3: Sum of carbon dioxide mole fraction and water vapor mole fraction  $\psi_{CO_2} + \psi_{H_2O}$  in test cases (a) *Air* and (b) *Oxy30* and normalized gas temperature according to test case *Air* (normalized by 1626 °C, compare figure 4.4 and figure 5.1(b)) and (c) soot and coal particle absorption coefficient ( $\kappa_s$ ,  $\kappa_c$ ) and (d) particle absorption coefficient of residual ash and fly ash ( $\kappa_{ra}$ ,  $\kappa_{fa}$ ; see section 4.2.3) against furnace width (in x-direction) from 0 to 4200 mm ( $1 \le i \le 42$ ) at the furnace center (distance to the wall in y-direction 2100 mm; j = 21) on furnace level 2 (distance to furnace top 2830 mm; k = 90), see figure 4.1 for orientation

emissivity model as it is in good agreement with EWBM results at low concentrations of carbon dioxide and water vapor, see figures 4.13(a) and 4.14(a) in section 4.3.2. Gas absorption coefficient of Oxy30 operation mode, however, is based on the WSGG4+1 model as it is in better agreement with EWBM data than the WSGG3+1 model at high concentrations of carbon dioxide and water vapor, see table 4.7 in section 4.3.2. In both cases, Patch mean absorption coefficient is applied, see section 4.3.4, and temperature and particle concentration are equal. It can be seen from figure 5.1(a) that gas absorption coefficient  $\kappa_g$  (here calculated as  $\kappa_{pa}$ ) is higher in Oxy30 than in Air due to higher gas emissivity. This, in turn, comes from higher concentration of radiating species in test case Oxy30. It can be seen from figure 5.2(a) that emissivity (at  $\Psi = 1$ ,  $\psi_{CO_2} = \psi_{H_2O}$ ) calculated from either WSGGM increases with increasing mole fraction of carbon dioxide and water vapor.

Figure 5.1 shows absorption coefficients at furnace level 2 (k = 90; 2830 mm from the furnace top) as function of furnace width. Gas absorption coefficient is on average 1.48 times higher for Oxy30. The figure shows that Oxy30 gas absorption coefficients are lowest at the furnace center. This results from lower gas emissivity at this position due to elevated temperatures. Gas emissivity decreases with increasing gas temperatures as shown in figure 5.2(b) for both emissivity correlations. In case of air firing, however, gas absorption coefficient does not decrease at the furnace center even though temperatures are higher in this region. This is due to the cosine shape of mole fraction distribution of carbon dioxide and water vapor assumed in both test cases. Normalized gas temperature and mole fraction of radiating species at furnace level 2 are plotted against furnace width in x-direction in figure 5.3 for both gas compositions. Mole fractions of radiating species are higher at the furnace center and compensate for the increase in temperature. This effect is stronger at low mole fractions of carbon dioxide and water vapor (as in Air) than at high mole fractions (as in Oxy30) since the slope of emissivity against mole fraction of radiating species is steeper at low concentrations, see figure 5.2(a). At higher values of carbon dioxide and water vapor concentration, further increase of concentration leads only to small increase in gas emissivity.

Figure 5.1(b) shows the ratio between gas absorption coefficient  $\kappa_g$  and particle absorption coefficient  $\kappa_{part}$ . The particle absorption coefficient is the sum of soot, coal, residual and fly ash absorption coefficients, see section 4.2.3:

$$\kappa_{part} = \kappa_s + \kappa_c + \kappa_{ra} + \kappa_{fa}. \tag{5.1}$$

Absorption coefficients of soot and coal are plotted against furnace width in x-

direction in figure 5.3(c) and absorption coefficient of residual and fly ash in figure 5.3(d) based on concentration assumptions made in section 4.2.3. At high coal and soot concentrations inside the flame, the ratio between gas and particle absorption coefficient becomes very low as shown in figure 5.1(b). Soot and coal particles dominate emission and absorption of heat radiation. Therefore, gas emissivity is of minor importance in these areas.

In the surrounding area of the flame, however, particle absorption coefficient is lower and gas absorption coefficient amounts to a larger proportion of the overall absorption coefficient. This corresponds with the findings of Krishnamoorthy et al. [72]. Near the furnace wall, where particle concentration is lower than in the flame region, the ratio between gas and particle absorption coefficient becomes > 1 under the assumptions made in section 4.2.3 with respect to oxy-fuel conditions (i.e., gas absorption coefficient is higher than particle absorption coefficient) while it remains < 1 in air firing, see also 5.1(b). Therefore, application of suitable emissivity models is advisable in order to yield results of similar quality as in the modeling of air-blown combustion.

Spectrally continuous emission and absorption of particles overlaps with wavenumber bands where gases emit and absorb heat radiation. Andersson et al.[4] report that radiative intensity can be lower by about 25 % in a dry lignite flame under oxy– fuel conditions if spectral overlap of particle and gas radiation is considered in their calculation. This means, that overall absorption coefficient is lower than the sum of the individual contributions of gas and particles. However, since influence of gas absorption coefficient appears to be low and consideration of spectral overlap further decreases its influence, the error introduced by neglecting overlap of particle and gas radiation is expected to be low with respect to radiative heat fluxes. Simulation results then imply maximum influence of elevated concentrations of carbon dioxide and water vapor on wall heat fluxes.

#### 5.1.2 Influence of Emissivity Correlations

This section describes the influence of the applied emissivity correlation on the gas absorption coefficient according to the flue gas composition in test case Oxy30. Section 4.3.4 shows how Patch absorption coefficient  $\kappa_{pa}$  [95] and effective absorption coefficient  $\kappa_e$  are calculated. The latter is commonly used in engineering calculations. The underlying gas emissivity model is either WSGG3+1 or WSGG4+1.



Figure 5.4: (a) Effective and (b) Patch gas absorption coefficient based on WSGG4+1 (solid line) and WSGG3+1 (dashed line) emissivity correlations according to flue gas composition in test case Oxy30 at the furnace center in y-direction (2100 mm from the wall, j = 21) of furnace level 3 (2830 mm from the furnace top, k = 90) against furnace width in x-direction

Figure 5.4(a) shows the effective absorption coefficients  $\kappa_e$  at the furnace center (2100 mm from the wall in y-direction, j = 21) of furnace level 3 (2830 mm from the furnace top, k = 90) against furnace width in x-direction from 0 to 4200 mm  $(1 \le i \le 42)$  based on either emissivity correlation. It can be seen from the figure that the effective absorption coefficient is almost equal in both cases. It is important to keep in mind that this result is related to the particular optical path length regarded here (L = 3.21 m). At higher optical path lengths, deviation between both effective absorption coefficients becomes larger, see figure 4.20.

Patch absorption coefficients based on either emissivity correlation are shown in figure 5.4(b). WSGG3+1 based Patch absorption coefficients are on average 2.6 times lower than the corresponding effective absorption coefficients. This factor is about 3.5 in case of WSGG4+1 based calculation. This result corresponds to the results shown in figure 4.20. Again, it is important to keep in mind that these results are only valid at L = 3.21 m. At optical path lengths > 5 m, deviation between Patch and effective gas absorption coefficient becomes much higher if based on WSGG3+1 emissivity. Therefore, combining WSGG3+1 based emissivity according to Smith et al. [122] and effective absorption coefficient should be avoided in case of high concentrations of carbon dioxide and water vapor and large optical path lengths.

WSGG3+1 based Patch absorption coefficients are approximately 1.3 times higher

than WSGG4+1 based Patch absorption coefficients. This means that the applied emissivity correlation is almost as influential as increased mole fractions of carbon dioxide and water vapor due to oxy–fuel combustion (which increases by a factor 1.48, see above in this section). Again, this is not a general result but only valid under the particular boundary conditions of this simulation. However, calculation of gas absorption coefficient by either Patch or effective method generally seems to have higher influence on resulting gas absorption coefficients than increased mole fractions of radiating species or emissivity correlation, especially at optical path lengths > 1 m.

## 5.2 Statistical Error



Figure 5.5: Normalized incident radiative heat flux against distance from the furnace top at the center of the furnace front wall (furnace center in x-direction, i = 22; at the wall in y-direction, j = 0) with  $q_{ref} = 162.2 \text{ kW/m}^2$  and (a) N = 1,233,520,000 and (b) N = 123,352,000

Producing results with low statistical error requires a sufficiently high number of beams being emitted from each control element. Monte Carlo results fluctuate around the correct solution since the method is a "repetition of tests on a physical model" [50]. Uncertainty can be decreased by increasing the number of tests (i.e., the number of emitted beams). One billion beams emitted from volume elements and 10,000 beams emitted from every surface element give a smooth distribution of radiative heat flux, see figure 5.5(a). The average number of beams emitted from each volume element is 4804. The total number of emitted beams is N = 1,233,520,000.

Furnace level		0	1	2	3	4	5	6
$egin{array}{c} z_d \ k \end{array}$	[mm] [-]	$\begin{array}{c} 700 \\ 112 \end{array}$	$\begin{array}{c} 1870 \\ 100 \end{array}$	$2830 \\ 90$	$\begin{array}{c} 4940 \\ 69 \end{array}$		$7940 \\ 39$	$9140 \\ 27$
$Measurement^1$	$[\mathrm{kW}/\mathrm{m}^2]$	33.3	53.0	53.9	68.7	63.0	62.3	49.5
$Deviation^2$	[%]	-19.1	-20.6	+5.5	+7.6	+14.9	-2.0	-0.1

**Table 5.1:** Relative deviation between modeled and measured radiative heat flux at the center of the furnace front wall (centered in x-direction, i = 22; at the wall in y-direction, j = 0), furnace levels 0 to 6 for test case Air

Radiative heat flux  $q_{rad}$  is normalized by a reference heat flux  $q_{ref} = 162.5 \text{ kW/m}^2$ in figure 5.5.

Reducing the number of beams by a factor of ten leads to fluctuation of radiative heat flux between neighbouring surface elements as can be seen in figure 5.5(b). Radiative heat fluxes remain within  $\pm 4\%$  of those based on simulation with ten times higher number of randomly emitted beams. Radiative heat flux in figure 5.5(a) still shows minor fluctuations between neighbouring surface elements attributed to the statistical error of Monte Carlo simulation. Although emitting more beams would lead to more accurate results, the number of beams is not increased further due to the required computing time.

# 5.3 Radiative Heat Flux

In this section, modeled radiative heat fluxes to the center of the furnace front wall are shown for test cases Air and Oxy30 in accordance with assumptions made in section 4.2. Modeled heat fluxes are then compared with measured values of incident radiative heat from section 3.2.4.

#### 5.3.1 Air Firing

Radiative transfer in test case Air is based on WSGG3+1 model [122] and Patch absorption coefficient. Figure 5.6 shows results of the simulation in comparison with measured values (estimated as shown in section 3.2.4). Error bars in figure 5.6 indicate the error range resulting from the standard deviation of the measured

 $<sup>^{1}</sup>$ Calculated from equation 3.4.

 $<sup>^{2}</sup>$ Between simulation (WSGG3+1 based, Patch absorption coefficient) and measurement.

Furnace level		0	1	2	3	4	5	6
${z_d \over k}$	[mm] [-]	$\begin{array}{c} 700 \\ 112 \end{array}$	$\begin{array}{c} 1870 \\ 100 \end{array}$	$2830 \\ 90$	$\begin{array}{c} 4940 \\ 69 \end{array}$		$7940 \\ 39$	$9140 \\ 27$
Measurement	$[\mathrm{kW}/\mathrm{m}^2]$	29.6	52.4	70.5	98.0	89.9	97.5	84.6
${ m Deviation}^3 { m Deviation}^4$	[%] [%]	$^{+8.1}_{+5.3}$	$^{+10.8}_{+9.8}$	$^{+22.4}_{+17.6}$	$^{+9.5}_{+4.6}$	$^{+15.8}_{+11.1}$	$-3.2 \\ -8.3$	$-1.6 \\ -9.4$

**Table 5.2:** Relative deviation between modeled and measured radiative heat flux at the center of the furnace front wall (centered in x-direction, i = 22; at the wall in y-direction, j = 0), levels 0 to 6 for test case Oxy30

average convective share  $\overline{\eta}$  which is described in section 3.2.4, see figure 3.5. Table 5.1 shows relative deviation between modeled and measured radiative heat transfer on levels 0 to 6.

Measured radiative heat fluxes are in good agreement with those obtained from simulation. Deviation of modeled values remains within  $\pm 20$  % of measured results. Deviation between both is highest in the near burner region where calculated incident radiative heat flux is lower than measured. This could be caused by slight underestimation of gas temperature by cosine shaped approximation as shown in section 4.2.1. However, good agreement between measured and calculated heat fluxes proves the general suitability of the modeling approach described in chapter 4 to predict incident radiative heat fluxes based on common global emissivity models. It also proves reliability of other assumptions (temperatures, particle concentration etc.).

#### 5.3.2 Oxy–Fuel

Radiative transfer by Monte Carlo simulation is calculated for test case Oxy30, see chapter 3, based on both WSGG3+1 and WSGG4+1 emissivity correlation. All other conditions are held constant. Patch absorption coefficient is used in the simulation for coupling emissivity model and RTE solution (see section 4.3.4). Figure 5.7 shows modeled radiative heat fluxes at the center of the furnace front wall against distance to the furnace top (centered in x-direction, i = 22; at the wall in y-direction, j = 0) in comparison with measured values.

 $<sup>^3\</sup>mathrm{Between}$  simulation (WSGG3+1 based, Patch absorption coefficient) and measurement.

<sup>&</sup>lt;sup>4</sup>Between simulation (WSGG4+1 based, Patch absorption coefficient) and measurement.



Figure 5.6: Measured (symbols) and calculated (solid line) radiative heat flux, case Air at the center of the furnace front wall



Figure 5.7: Measured (symbols) and modeled radiative heat flux of test case Oxy30 based on WSGG3+1 (dashed line) and WSGG4+1 (solid line) emissivity correlation at the center of the furnace front wall

Both simulations are in good agreement with the measurement. Relative deviation between measured and modeled values is shown in table 5.2. WSGG4+1 based modeling produces slightly better agreement (average deviation 9.4 %) with measured radiative heat fluxes compared to WSGG3+1 based simulation (average deviation 10.2 %) except for level 5 and 6. While heat flux results on other levels are overestimated, both simulations underrate radiative heat transfer to the walls at these levels. This might be related to inappropriate assumptions on temperature distribution downstream of the over fire air ports or not considering the flue gas outlet to the second boiler pass in the simulation.

Figure 5.7 shows that resulting heat fluxes are very similar in both WSGG3+1 and WSGG4+1 based simulation. WSGG4+1 modeling results are on average about 5 % lower than WSGG3+1 based heat flux. The reason is that WSGG3+1 based absorption coefficients are approximately 1.3 times higher than those based on WSGG4+1 emissivity data, see figure 5.4(b). WSGG4+1 emissivities are about 10 % lower than that calculated from WSGG3+1 under oxy-fuel conditions at the respective path length of 3.21 m, see figure 4.16. Deviation between both simulations is highest (about 10 %) in regions of high temperatures around  $z_d = 5000$  mm which comes from increasing deviation between WSGG3+1 and WSGG4+1 emissivities at elevated gas temperatures as illustrated in figure 4.16(b) and 4.17(b). Altogether, the emissivity correlation used in the simulation is of minor importance with respect to heat flux results. Application of WSGG3+1 generated emissivities [122] leads to results of similar accuracy as the WSGG4+1 based simulation under the given conditions.

## 5.4 Modeling Parameters and Their Influence

Several quantities influence the modeling of radiative heat transfer in furnaces. In the following sections, some of these are investigated in more detail in order to understand their influence on radiative heat flux. The impact of flue gas composition of oxy-fuel combustion is compared to that of nitrogen-diluted flue gas from combustion with air under the assumption of equal temperature distribution in both cases. Influence of gas temperature is analysed by modeling operation modes with different oxygen concentration in the oxidant gas and comparing the heat flux results with measured incident radiative heat fluxes. Furthermore, inhomogeneous flue gas composition in the furnace and coupling of emissivity model and RTE by either effective or Patch absorption coefficient is analysed. Finally, influence of wall reflectivity on predicted heat fluxes is investigated.

#### 5.4.1 Flue Gas Composition

Comparing figure 5.6 and figure 5.7 in section 5.3 shows that incident radiative heat flux at the furnace front wall is on average 30 % (between 7 % and 50 %) higher in Oxy30 than in Air. This is, however, mainly a result of higher gas temperatures in Oxy30 and does not provide direct information on the influence of increased concentration of carbon dioxide and water vapor.

Influence of radiative species concentration on heat flux can be seen from figure 5.8 where results of three different simulations based on equal distribution of particles (based on the assumptions made for test case Oxy30) and gas temperature (based on assumptions made for test case Air)<sup>5</sup> are presented. Mole fractions of carbon dioxide and water vapor correspond to the respective operation mode, either Air or Oxy30. Gas emissivity is calculated either from WSGG3+1 (air firing) or WSGG4+1 (oxy-fuel) model, respectively. The third calculation is based on non-radiating gas atmosphere ( $\kappa_g = 0$ ) in order to estimate the contribution of coal, ash and soot particles to radiative transfer.

Figure 5.8 shows radiative heat fluxes at the center of the furnace front wall (at j = 0, 2100 mm distance from the wall in x-direction, i = 22) against distance from the furnace top for the three simulations. Incident radiative heat flux is on average 7.3 % higher in air-firing than in oxy-fuel combustion with respect to flue gas composition. This is a direct result of increased gas emissivity and indicates that increased concentration of radiating species resulting from oxy-fuel combustion contributes to slightly higher heat fluxes provided that temperatures are equal and both emissivity correlations produce accurate results. For example, emissivity of a gas mixture with  $\psi_{CO_2} = 0.15$ ,  $\psi_{H_2O} = 0.1$  and  $T_g = 1500$  K at L = 3.21 m is 0.327 and, thus, approximately 30 % lower than gas emissivity of a gas mixture with  $\psi_{CO_2} = 0.25$  which is 0.429, see also figure 4.12.

Although gas emissivity increases by about 30 %, increase in radiative heat flux is only 7.3 %. The reason is that gas radiation contributes only to a certain proportion to radiative transfer inside the furnace as can be seen from figure 5.8. Assumption of non-radiating gas mixture (i.e., only particle radiation is considered) leads to

<sup>&</sup>lt;sup>5</sup>There is no particular reason to choose this combination other than to enable a comparison independent from temperature and particle distribution.



Figure 5.8: Radiative heat flux at the center of the furnace front wall for different gas atmospheres: air firing (solid line), oxy-fuel combustion (dashed line) and non-radiating gas mixture (dotted line) calculated from simulations based on equal temperature and particle distribution

radiative heat fluxes in the order of 70 to 90 % of heat fluxes based on air-firing flue gas composition. Thus, gas radiation accounts only for approximately 10 to 30 % of total radiative heat flux. Share of gas radiation is smaller at the furnace top (near the burner) since particle concentration there is higher. These results are in basic agreement with findings of Andersson et al. [4]. In their study, the share of gas radiation is 34 % or more at the wall of an oxy-fuel furnace depending on oxygen concentration in the oxidant gas. In air-blown combustion, the share of gas radiation is about 30 % in their study.

#### 5.4.2 Soot Concentration and Coal Particle Size

A maximum soot concentration of  $f_{v,s,0} = 10^{-6}$  is assumed in the calculation, see section 4.2.3. Figure 5.9 shows heat flux results of test case Oxy30 based on this assumption (solid line, "Mean"). In order to investigate sensitivity of heat flux results towards the assumption made on soot concentration, calculation based on lower soot concentration  $f_{v,s,0} = 10^{-9}$  is performed and heat flux results are presented in figure 5.9 (dashed line, "Low Soot"). It becomes clear from the figure that the assumption made on soot concentration has only minor influence on heat flux prediction within the range  $f_{v,s,0} = 10^{-9}$  to  $10^{-6}$  [74].



Figure 5.9: Radiative heat flux at the center of the furnace front wall, test case Oxy30, based on different assumptions on soot concentration and average coal particle size: soot concentration  $f_{v,s,0} = 10^{-6}$  and average particle size diameter  $d_{c,m} = 82.5 \cdot 10^{-6}$  m (curve "Mean"), soot concentration  $f_{v,s,0} = 10^{-9}$  and average particle size diameter  $d_{c,m} = 82.5 \cdot 10^{-6}$  m (curve "Low Soot"), and soot concentration  $f_{v,s,0} = 10^{-6}$  and Sauter particle size diameter  $d_{c,s} = 121.3 \cdot 10^{-6}$  m (curve "Sauter")

An average diameter of coal particles  $d_{c,m} = 82.5 \cdot 10^{-6}$  m is assumed in the modeling according to the particle size analysis presented in table 4.4, section 4.2.3. However, it is also possible to find other average diameters such as Sauter diameter, see section 4.2.3. Figure 5.9 shows heat flux results of test case Oxy30 based on both, average diameter  $d_{c,m}$  (solid line, "Mean") and Sauter diameter  $d_{c,s} = 121.3 \cdot 10^{-6}$  m (dotted line, "Sauter"). It can be seen from the figure that heat flux results are similar under both assumptions. Heat flux results based on Sauter diameter are slightly higher close to the furnace top and lower at lower furnace elevations than those based on average particle diameter  $d_{c,m}$ . However, agreement between predicted and measured heat fluxes is very similar in both cases.

#### 5.4.3 Gas Temperature

Gas temperature increases with increasing oxygen concentration in the oxidant gas as can be seen from measurements presented in chapter 3. Oxygen concentration



Figure 5.10: (a) Normalized radiative heat received by the surface elements against oxygen concentration in the oxidant  $y_{O_2}$  (×-symbols) and normalized radiative heat received by the surface elements for case Air (dotted line); (b) relative deviation of radiative heat received by the surface elements between WSGG3+1 and WSGG4+1 based simulation

increases if the mass flow of recirculated flue gas decreases and the mass flow of oxygen remains constant. If the amount of flue gas and, hence, overall heat capacity decreases, temperature increases.

Figure 5.10(a) shows the total radiative heat (normalized by 20 MW) received by the surface elements of the enclosure (i.e., the furnace walls) in operation cases Oxy24, Oxy28, Oxy32 and Oxy36 against oxygen concentration in the oxidant gas. It can be seen from the figure that radiative heat received by surface elements increases with increasing oxygen concentration in the oxidant gas. The dotted line shows normalized radiative heat received by surface elements in test case Air. Radiative heat received by the enclosure walls is similar in Oxy28 and Air. This indicates that an oxygen concentration of approximately 28 % by volume (wet) results in radiative heat uptake similar to that of air-blown combustion. Other authors also identified this oxygen concentration to result in similar heat transfer compared to air-blown combustion [84].

Figure 5.10(b) shows the relative deviation between WSGG3+1 based calculation of radiative heat received by all surface elements and corresponding values of WSGG4+1 based simulation. WSGG3+1 based modeling leads to higher values but deviation remains below 6 % in all test cases and increases with decreasing oxygen concentration.



Figure 5.11: Incident radiative heat flux at the center of the furnace front wall at different oxygen concentrations in the oxidant gas: comparison between measurement and simulation results based on WSGG3+1 and WSGG4+1

Appropriate setting of oxygen concentration in the oxidant gas flow possibly yields similar radiative heat transfer in oxy-fuel as in air-blown combustion. Therefore, it is also easy to compensate for higher gas emissivity under oxy-fuel conditions by adjusting the oxygen concentration. Figure 5.11 shows measured radiative heat fluxes in test cases Oxy24, Oxy28, Oxy32 and Oxy36 in comparison with calculated values based on the Patch absorption coefficient. It becomes clear from the figure that deviation between WSGG3+1 and WSGG4+1 based simulation is rather small in comparison with general deviations between measured and modeled values. Application of either WSGG4+1 or WSGG3+1 gas emissivity model seems to have a negligible effect on radiative heat fluxes with respect to optical path length and flue gas composition in this particular case.



Figure 5.12: Incident radiative heat flux at the center of the furnace front wall, test case Oxy30, based on simulations with 30 % higher gas temperature  $(1.3 \times T_g)$  and 30 % higher gas absorption coefficient  $(1.3 \times \kappa_g)$ 

By contrast, gas temperature has a significant effect on calculated radiative heat fluxes. In section 5.4.1, figure 5.8 shows increased incident radiative heat fluxes resulting from increased gas emissivity of flue gas under oxy-fuel conditions. Flue gas emissivity is approximately 30 % higher compared to flue gas of air-blown combustion. In contrast hereto, 30 % increase in gas temperature results in significant increase of incident radiative heat flux at the furnace front wall, see figure 5.12. Radiative heat fluxes are approximately three times higher since energy content of each beam is a function of the fourth power of gas temperature, equation 4.3, while it is only a linear function of gas absorption coefficient. Figure 5.12 also shows resulting incident radiative heat fluxes at the furnace front wall if gas absorption coefficient is multiplied by a factor of 1.3. In that case, resulting heat fluxes are on average 4 % higher. It becomes clear from the figure that optical gas properties are of minor significance compared with gas temperature.

### 5.4.4 Concentration Profiles

Gas emissivity of each volume element is calculated according to local concentrations of carbon dioxide and water vapor. Concentration of both species is constant within each volume element and calculated from equation 4.27 in section 4.2.2. Figure



Figure 5.13: (a) Relative deviation of calculated incident radiative heat fluxes of test case Oxy30 at the center of the furnace front wall against distance to the furnace top based on constant mole fractions  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  towards simulation based on cosine-shaped profiles of carbon dioxide and water vapor mole fraction (equation 4.27 in section 4.2.2); (b) mole fraction ratio  $\Psi$  in test case Oxy30 at furnace level 1 (1870 mm from the furnace top, centered in y-direction) if based on cosine mole fraction profiles according to equation 4.27 (solid line) or constant mole fractions  $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$  (dashed line) against furnace width in x-direction (see figure 4.1 for orientation)



Figure 5.14: Emissivity of flue gas with  $\psi_{CO_2} + \psi_{H_2O} = 0.95$  at T = 1000 K and L = 3.21 m as function of mole fraction ratio  $\Psi = \psi_{H_2O}/\psi_{CO_2}$ calculated from WSGG4+1 emissivity correlation

#### (b) Mole fraction ratio

5.13(a) shows the

relative deviation in 
$$[\%] = 100\% \cdot \frac{q_{rad}^* - q_{rad}}{q_{rad}}$$
 (5.2)

between two different simulations based on test case Oxy30: concentration profile of both gas species calculated according to equation 4.27 ( $q_{rad}$  in equation 5.2) and concentration of both species assumed to be constant (homogeneously distributed) in *all* volume elements ( $\psi_{CO_2} = 0.7$  and  $\psi_{H_2O} = 0.25$ ,  $q_{rad}^*$  in equation 5.2).

Figure 5.13(b) shows the mole fraction ratio  $\Psi = \psi_{H_2O}/\psi_{CO_2}$  at the center of furnace level 1 (distance from furnace top 1870 mm, k = 100; distance from the wall in y-direction 2100 mm, j = 21) in both cases. Mole fraction ratio  $\Psi$  is 0.357 and hence lower in case of homogeneous (constant) distribution of carbon dioxide and water vapor concentration. Its constant value 0.357 lies in the interval 0.231  $\leq$  $\Psi \leq 0.407$  of the WSGG4+1 spline function representing weights  $a_{\epsilon,u}(T, \Psi)$  and absorption coefficients  $\kappa_u(\Psi)$ , see section 4.3.2. Mole fraction ratios resulting from cosine shaped concentration profiles are within the interval 0.407  $\leq \Psi \leq 0.759$  of this function. Apparently, both calculations yield similar results. Although gray solution of the RTE actually requires homogeneous media [76], deviation in radiative heat flux is less than 5 % and mainly due to statistical error of Monte Carlo simulation. The assumption made on flue gas composition does not show significant influence on the simulation results.

Figure 5.14 shows WSGG4+1 gas emissivities of a gas mixture with carbon dioxide, water vapor and 5 % non-radiating species ( $\psi_{CO_2} + \psi_{H_2O} = 0.95$ ) as function of mole fraction ratio  $\Psi$ . The figure illustrates that slight changes of mole fraction ratio only lead to minor changes of gas emissivity (e.g., between  $\Psi = 0.35$  and 0.5, gas emissivity increases only by approximately 6 %). At low values of  $\Psi$ , the curve shows highest gradients.

Altogether, both assumptions on flue gas composition (i.e., cosine profile or constant values) lead to similar results with respect to incident radiative heat fluxes. High concentration gradients of radiating gas species occur especially inside the flame where radiation by particles dominates. If the purpose of the simulation is to find wall heat fluxes, assuming homogeneous distribution of gas species concentration leads to satisfactory results. Assumption of cosine shaped concentration profiles of radiating gas species apparently neither significantly improves nor worsens the results.

### 5.4.5 Gray Absorption Coefficient

Either Patch absorption coefficient  $\kappa_{pa}$  [95] or effective absorption coefficient  $\kappa_e$ , see figure 4.20 in section 4.3.4, are used in the simulation. If absorption and emission by particles is neglected, it can be shown for oxy-fuel combustion that radiative heat fluxes are significantly higher (approximately 150 %) if the effective absorption coefficient  $\kappa_e$  is used instead of  $\kappa_{pa}$ , see figure 5.15.

If particle radiation is included in the model, influence of gas absorption and emission on wall heat fluxes decreases. The dashed line in figure 5.16 shows heat flux results of calculation based on  $\kappa_{pa}$  and the dotted line shows results of  $\kappa_e$  based simulation. The latter yields significantly overrated radiative heat fluxes compared with measured values whereas simulation based on Patch absorption coefficient yields results which are in reasonable agreement with measured values.

However, differences in heat flux results would be smaller between both simulations if temperatures were unknown and iterative solution of the RTE was required. In this case an assumption is made on temperature distribution and the RTE is solved accordingly. From energy balances of each volume element, new temperatures are calculated and the RTE is solved again. This procedure repeats until temperatures converge to stable values. The applied absorption coefficient then influences both, heat flux and temperature. Calculations in chapter 6 are based on an iterative solution approach of the RTE. Influence of gas absorption coefficient on radiative heat flux is there lower compared to the non–iterative calculation where temperatures are fixed, see also table 1.1 in section 1.3.

Comparing figure 5.16 and figure 5.7 reveals that the error introduced by Smith's WSGG3+1 emissivity correlation is small compared to the application of  $\kappa_e$  as mean absorption coefficient. From figure 4.20 it can be concluded that combined use of Smith's WSGG3+1 and effective absorption coefficient  $\kappa_e$  overrates gas optical properties even more at path lengths > 5 m. This possibly leads to overrated incident heat fluxes and underestimation of gas temperature.

#### 5.4.6 Wall Properties

In gray Monte Carlo simulation, furnace walls are gray which means that absorption and emission of heat radiation is independent of wavelength, absorptivity and emissivity of the wall are equal and absorptivity and emissivity are < 1 (i.e., a certain



Figure 5.15: Radiative heat flux at the center of the furnace front wall for Oxy30without particle radiation ( $\kappa_{part} = 0$ ) using the Patch absorption coefficient  $\kappa_{pa}$  (solid line) and the effective absorption coefficient  $\kappa_{e}$ (dashed line); gas emissivity calculated from WSGG4+1



Figure 5.16: Radiative heat flux at the center of the furnace front wall for Oxy30 based on the Patch absorption coefficient  $\kappa_{pa}$  (solid line) and the effective absorption coefficient  $\kappa_e$  (dashed line); gas emissivity calculated from WSGG4+1



Figure 5.17: Relative deviation between modeled incident radiative heat fluxes at the center of the furnace front wall calculated from heat transfer simulation with  $\rho_w = 0.25$  and  $\rho_w = 0.15$  plotted against distance from the furnace top

share of incident radiation is reflected). In coal-fired furnaces, wall reflectivity depends on optical properties of ash layers formed at the wall surface. Wall reflectivity  $\rho_w$  is assumed to be 0.15 in all simulations.

High reflectivity leads to less absorption of heat radiation. Reflected radiation is either absorbed by flue gas or reaches another wall where it is either absorbed or reflected again. Probability of absorption at another wall also decreases with increasing wall reflectivity, see equation 4.16 in section 4.1.3. Thus, heat uptake by furnace walls decreases with increasing wall reflectivity and furnace exit flue gas temperature increases.

Since flue gas temperatures known from measurements are approximated as shown in section 4.2.1, the assumption on wall reflectivity has only little impact on heat flux results. If wall reflectivity is assumed to be high in the model, the amount of incident radiation being reflected by the walls increases. The energy content of a reflected beam is then absorbed by flue gas (if the reflected beam does not arrive at another wall prior to absorption) and contributes to the internal energy of the control element and, hence, leads to an increase in temperature. In the simulation, by contrast, it does not affect temperatures since they are pre-defined and the solution is non-iterative. Therefore, the assumption on wall reflectivity is of minor significance here. This, however, should not be mixed up with the important influence of wall reflectivity on heat transfer in furnaces.

It has to be kept in mind also that the incident radiative heat flux is measured by means of a hollow ellipsoidal radiometer, see section 3.2.1. The radiometer absorbs all incident radiation. Thus, measured heat flux values have to be compared to total incident radiative heat flux including the share which is actually reflected at the wall.

Figure 5.17 shows relative deviation between radiative heat flux results if  $\rho_w = 0.25$ and results obtained if  $\rho_w = 0.15$ . All other parameters are based on test case Oxy30. Maximum deviation of simulation results is approximately 8 % at the top of the furnace.

# 6 Application

Simulation of radiative heat transfer as described in chapter 4 and the results shown in chapter 5 serve as calibration of mean absorption coefficient and gray emissivity. Accuracy is checked by comparing measured and calculated radiative heat fluxes. Gas temperature is based on measured values. However, temperature distribution in furnaces is usually unknown and has to be calculated. Therefore, an iterative solution of the RTE is required where temperatures are calculated from heat balances at each volume element, see table 1.1 in section 1.3. As a result, gas absorption influences all governing processes including flow and chemical reactions. In order to evaluate the effect of gas absorption coefficient on the results of an iterative solution, WSGG4+1 correlation from section 4.3.2 is implemented in the commercial CFD code FLUENT 13.0 and heat transfer inside the test facility described in section 3.1 is calculated.

# 6.1 CFD Modeling

By default, Smith's WSGG3+1 emissivity correlation is applied in the CFD solver and gas absorption coefficient is calculated as effective absorption coefficient, see table 2.1 in section 2.2.4. The WSGG4+1 model presented in section 2.2.2 is implemented as user-defined function (UDF) to allow for high carbon dioxide and water vapor concentrations in the flue gas. The UDF is then applied in the simulation of heat transfer in the test furnace described in section 3.1.

The RTE is solved by the discrete ordinates method, see references [37–39]. The discrete ordinates method solves the RTE for a finite number of directions in the Cartesian coordinate system. Each direction represents a discrete solid angle in the whole solid angle of  $4\pi$ . All discrete solid angles are characterized by the same polar and azimuthal control angles  $\Delta\theta$  and  $\Delta\phi$ , respectively [41]. Each octant with respect to the Cartesian coordinate system is divided into four discrete solid angles resulting in 32 directions in total.

Furnace walls are assumed to emit gray and diffusely (i.e., the diffuse fraction  $f_d$  [41] is 1 in the calculation) and reflection is assumed to be diffuse as well. Wall reflectivity  $\rho_w$  is constant.

Multiphase flows (gas and particles) are calculated as stationary and incompressible with Eulerian gas phase and Lagrangian particle phase including friction and gravity. Turbulence is modeled with the realizable  $k-\epsilon$ -model of Shih et al. [117]. The particle phase represents coal and ash particles inside the furnace.<sup>1</sup> Coal and ash particles are assigned a constant emissivity  $\epsilon_{part}$ . If coal and ash particles are included in the calculation, scattering by soot particles is neglected [41]. Soot absorption coefficient  $\kappa_s$  is calculated from

$$\kappa_s = b_1 \rho_s \left( 1 + b_T \left( T_s - 2000 \right) \right) \tag{6.1}$$

with  $\rho_s$  being the mass concentration of soot in the flue gas,  $T_s$  its temperature, and the constants  $b_1 = 1232.4 \text{ m}^2/\text{kg}$  and  $b_T \approx 4.8 \cdot 10^{-4} \text{ K}^{-1}$  [41]. The sum of soot absorption coefficient  $\kappa_s$  and gas absorption coefficient  $\kappa_g$  is then assumed to be the absorption coefficient of the gas–soot–mixture  $\kappa_{g+s}$  [41]. Calculation of coal and ash radiative properties is described in section 6.2.2.

Pyrolysis of solid fuel particles is modeled by the two competing rates model of Kobayashi et al. [69, 70]. The model considers two weighted temperature dependent devolatilization rates which represent release of volatiles at different temperature ranges:

$$R_1 = B_1 \exp\left(\frac{-E_1}{R T_{part}}\right) \tag{6.2}$$

$$R_2 = B_2 \exp\left(\frac{-E_2}{R T_{part}}\right) \tag{6.3}$$

with  $R_1$  and  $R_2$  being the two devolatilization rates,  $B_1$  and  $B_2$  pre-exponential factors of the Arrhenius form and  $E_1$  and  $E_2$  respective values of activation energy. Volatile release is expressed as a function of time by [69]

$$\frac{m_{vol}(t)}{m_{part,0} - m_{ash}} = \int_{0}^{t} (\alpha_1 R_1 + \alpha_2 R_2) \exp\left(-\int_{0}^{t} (R_1 + R_2) dt\right) dt$$
(6.4)

where  $m_{vol}(t)$  is the mass of volatiles released from the particle at time t,  $m_{part,0}$  is the initial water-free mass of the particle,  $m_{ash}$  is the mass fraction of ash in the particle and  $\alpha_1$  and  $\alpha_2$  are the weighting factors. One of the weighting factors is

<sup>&</sup>lt;sup>1</sup>Particle phase does not include soot particles.

assumed to be "the [mass] fraction of volatiles [of the fuel] determined by proximate analysis" [107] since it "represents devolatilization at low temperature" [41]. The other weighting factor is set to unity as it represents "the yield of volatiles at very high temperature" [107].

Char oxidation is modeled by the surface reaction intrinsic model, cf. reference [121]. In this model "the surface reaction rate includes the effects of both bulk diffusion and chemical reaction" [41] and the order of reaction is assumed to be equal to unity [41]. Char combustion rate is calculated from the expression:

$$\frac{dm_{part}}{dt} = -A_{part} \psi_{O_2} \left(\frac{D_0 R_{int}}{D_0 + R_{int}}\right)$$
(6.5)

where  $m_{part}$  is the particle mass,  $A_{part}$  its surface area and  $\psi_{O_2}$  the local oxygen mole fraction.  $D_0$  is the diffusion rate coefficient taken from the model of Field [36] and  $R_{int}$  is the intrinsic reaction rate which "incorporates the effects of chemical reaction on the internal surface of the char particle [...] and pore diffusion" [41]. It is calculated from [41]

$$R_{int} = \eta_{int} \frac{d_{part}}{6} \rho_{part} A_{int} k_{int}$$
(6.6)

where  $\eta_{int}$  is "the ratio of the actual combustion rate to the rate attainable [...] if no pore diffusion resistance existed" [121] also referred to as effectiveness factor, cf. reference [78],  $d_{part}$  is the particle diameter,  $\rho_{part}$  its density and  $A_{int}$  its specific internal surface area which is assumed to be constant during char combustion. Values of  $A_{int}$  of chars are provided in reference [120]. The intrinsic reaction rate  $k_{int}$  is of Arrhenius form [41]

$$k_{int} = B_{int} \, \exp\left(\frac{-E_{int}}{R \, T_{part}}\right). \tag{6.7}$$

Values of  $B_{int}$  and  $E_{int}$  for chars can be found in reference [120]. Particle diameter as function of time  $d_{part}(t)$  is calculated from

$$d_{part}(t) = d_{part,0} \left(\frac{m_{part}(t)}{m_{part,0}}\right)^{\frac{1}{4}}$$
(6.8)

with  $d_{part,0}$  being the initial particle diameter and  $m_{part,0}$  the initial particle mass. The exponent 1/4 allows for decrease of both, particle size and particle density with increasing t [41]. In oxy-fuel atmosphere, the program's multiphase surface reactions model is used so that besides oxidation other heterogenous reactions as Boudouard and watergas reaction can be included, see reference [41] for details.

# 6.2 Results

Several cases with different radiative properties of the flue gas are investigated using the FLUENT software. Parameters under study are gas absorption coefficient (either Patch absorption coefficient  $\kappa_{pa}$  or effective absorption coefficient  $\kappa_e$ ), particle emissivity and wall reflectivity.

At first, particles are assigned an emissivity of zero (i.e., non-participating particle phase) in order to identify the sole effect of gas radiation, section 6.2.1. Then, particle emissivity is set to 0.7 and 0.9 in order to investigate the influence of gas optical properties if particle radiation dominates radiative transfer, section 6.2.2. Lockwood and coworkers [87] use linear equations of the local mass fraction of unburnt char implying an emissivity of 1.0 for coal and 0.6 for ash. Since emissivity of particles is constant (not depending on combustion progress) in the FLUENT calculation, aforesaid constant values are used as "average" emissivity of coal and ash particles.

WSGG4+1 emissivity correlation is used in each of the cases shown in table 6.1. Calculation of Patch absorption coefficient is implemented as additional UDF in FLUENT according to equation 4.61. Each test case is part of an univariant sensitivity analysis addressing either gas absorption coefficient, particle emissivity or wall reflectivity.

Case No.	$\kappa_g$	$\epsilon_g$	$\epsilon_{part}$	$ ho_w$
1	Effective	WSGG4+1	0.0	0.3
2	Patch	WSGG4+1	0.0	0.3
3	Effective	WSGG4+1	0.9	0.3
4	Patch	WSGG4+1	0.9	0.3
5	Effective	WSGG4+1	0.7	0.3
6	Patch	WSGG4+1	0.7	0.3
7	Patch	WSGG4+1	0.7	0.15

**Table 6.1:** Test cases and the assumptions made on gas absorption coefficient  $\kappa_g$ , gas emissivity  $\epsilon_g$ , particle emissivity  $\epsilon_{part}$  and wall reflectivity  $\rho_w$  in each case



Figure 6.1: Gas absorption coefficient  $[m^{-1}]$  at the center cross-section of the furnace (case 1 and case 2 in table 6.1)

### 6.2.1 Gas Atmosphere with Non–Participating Particles

As shown in section 4.3.4, effective and Patch absorption coefficient may become very different from each other. Depending on gas composition and optical path length, the effective absorption coefficient becomes significantly higher than the Patch absorption coefficient. Figure 6.1 corresponds to this finding: figure 6.1(a) and figure 6.1(b) show effective and Patch based absorption coefficient values and their distribution in the furnace. Particle emissivity is assumed to be zero in both cases. Comparing both figures shows that Patch absorption coefficient is on average five times lower. This is basically in accordance with figure 4.20 in section 4.3.4. It can be seen from the figure that the gas absorption coefficient is lower at the furnace center. This is due to higher gas temperatures in that region. Gas emissivity decreases with increasing temperature due to decreasing density, see figure 5.2.

Figure 6.2 shows gas temperatures in both cases. It can be seen from this figure that gas temperatures are higher at the furnace center. Gas temperature is overrated in comparison with measured values (not shown here). The simulation yields temperature values > 1400 °C while measured values are < 1300 °C, see figure 4.5 in section 4.2.1. Temperatures in case 2, figure 6.2(b), are somewhat higher than those in case 1, figure 6.2(a). This is because Patch absorption coefficient is lower than the effective absorption coefficient. Differences in gas temperature of both cases are



Figure 6.2: Gas temperatures  $[^{\circ}C]$  at the center cross–section of the furnace (case 1 and case 2 in table 6.1)



Figure 6.3: Incident radiative heat flux  $\rm [kW/m^2]$  at the furnace front wall (case 1 and case 2 in table 6.1)

#### < 100 K.

Incident radiative heat fluxes at the front wall are shown in figure 6.3. Heat fluxes are significantly lower in case 2 due to lower Patch gas absorption coefficient and despite higher gas temperatures. Maximum heat flux in case 1 is between 122 and 129 kW/m<sup>2</sup> while it is only between 76 and 84 kW/m<sup>2</sup> in case 2. This tendency is in accordance with results of Monte Carlo simulation shown in figure 5.15 in section 5.4.5. Radiative heat flux increases with increasing gas absorption coefficient.

FLUENT and Monte Carlo results yield different incident radiative heat fluxes at the furnace walls. Monte Carlo simulation based on effective absorption coefficient, figure 5.15, shows maximum values at about 100 kW/m<sup>2</sup> and Patch based maximum values are approximately 36 kW/m<sup>2</sup>. In both cases, values of incident radiative heat flux are lower in comparison with FLUENT results. Since overall absorption coefficient is underestimated by the assumption of non-participating particles, the iterative solution in FLUENT overrates gas temperature which, in turn, leads to high incident radiative heat fluxes. Therefore, FLUENT results on incident radiative heat flux are higher than those calculated from fixed gas temperatures in the Monte Carlo simulation.

## 6.2.2 Gas Atmosphere with Participating Particles

The model includes radiatively participating particles with their emissivity being fixed at 0.9 in test cases 3 and 4. As a result, the absorption coefficient of the gas-particle-mixture is higher in comparison with case 1 and 2. Gas absorption coefficients are either modeled as effective, figure 6.4(a), or Patch absorption coefficient, figure 6.4(b). Again, the effective gas absorption coefficient of case 3 turns out to be higher than the Patch gas absorption coefficient of case 4, see section 4.3.4, figure 4.20.

Gas absorption coefficient is essentially the same in both cases, figures 6.1 and 6.4. There are only minor differences due to different gas temperatures since including particle radiation results in lower temperatures. Gas absorption coefficient becomes slightly higher due to higher gas temperature, see figure 5.2(b).

Gas temperature distribution in case 3 and 4 at the furnace center is shown in figure 6.5. In both cases, gas temperatures are lower compared with cases 1 and 2, see figure 6.2. Moreover, gas temperature distribution of both cases is similar in contrast to case 1 and 2. This is a result of including particle radiation. Emissivity and, hence,



Figure 6.4: Gas absorption coefficient  $[m^{-1}]$  at the center cross-section of the furnace (case 3 and case 4 in table 6.1)

absorption coefficient become significantly higher due to the spectrally continuous radiation. The particle absorption coefficient is much higher than the absorption coefficient of the gas. Therefore, the overall absorption coefficient (i.e., the sum of gas and particle absorption coefficient) is almost independent of gas absorption coefficient.

However, gas absorption coefficient has a small effect on incident radiative heat flux. Radiative flux at the front wall is lower in case 4 than in case 3, see figure 6.6. This result is in basic accordance with the findings of section 5.4.5, figure 5.16, showing that application of the effective gas absorption coefficient leads to overprediction of radiative heat flux. However, both FLUENT calculations are more similar to each other than the corresponding results of the Monte Carlo simulation in section 5.4.5.

Overprediction of radiative heat flux results from overpredicted temperatures. Figures 6.7 and 6.8 show measured and calculated gas temperatures on level 1 and 6, respectively. It can be seen from the figures that the agreement between measurement and modeling results is good on level 1. However, FLUENT simulation overpredicts gas temperatures at the center of furnace level 6 where cosine shaped approximation according to section 4.2.1 is still in good agreement. Figure 6.9 compares measured and modeled incident radiative heat fluxes at the center of the furnace front wall. It can be seen from the figure that heat fluxes are overpredicted by the simulation.



Figure 6.5: Gas temperatures  $[^{\circ}C]$  at the center cross-section of the furnace (case 3 and case 4 in table 6.1)



Figure 6.6: Incident radiative heat flux  $[kW/m^2]$  at the furnace front wall (case 3 and case 4 in table 6.1)

Since simulation comprises a number of boundary conditions and models which affect each other, reasons for overprediction of radiative heat fluxes and gas temperatures are difficult to identify. Since temperatures are partly overrated, furnace wall reflectivity might be lower than the assumed value of 0.3. In order to investigate the results' sensitivity towards wall reflectivity, a value of 0.15 is assumed in case 7. If  $\rho_w$  decreases, more heat radiation can be absorbed by the walls leading to lower gas temperatures and, hence, incident radiative heat fluxes.

Gas optical properties are unlikely to cause deviation between simulation results and measured values: on the one hand, overprediction of heat fluxes at the wall occurs for all cases and, on the other hand, gas absorption coefficient is rather small compared to the overall absorption coefficient of the gas-particle-mixture. Figure 6.10 illustrates the ratio between gas absorption coefficient and (coal and ash) particle absorption coefficient in case 3 and 4. It is between 0.133 and 0.2 in case 3 and < 0.067 in case 4. Soot particles are considered by a separate approach described in section 6.1.

Although considering the contribution by soot particles, particle absorption coefficients calculated in section 4.2.3 are lower<sup>2</sup> than absorption coefficients calculated according to section 6.1 where the solid phase includes only coal and ash particles. Results of section 4.2.3 are in better agreement with values published by Krishnamoorthy et al. [71, 72]. They predict ratios between gas and particle absorption coefficients > 1 at some positions in a coal-fired oxy-fuel boiler. This leads to the conclusion that coal and ash particle absorption coefficient in the FLUENT model is too high and possibly leads to overprediction of incident radiative heat flux.

Absorption coefficient of coal and ash particles<sup>3</sup> is calculated according to the following equation [41]:

$$\kappa_{part} = \sum_{n=1}^{N} \epsilon_{part,n} \frac{A_{part,n}}{V}$$
(6.9)

with  $\epsilon_{part,n}$  and  $A_{part,n}$  being emissivity and projected area of N particles in the control volume V, respectively. Particle emissivity is assumed to be 0.9 in the simulation of case 3 and 4. The projected area of particles is calculated from

$$A_{part} = \frac{\pi d_{part,n}^2}{4} \tag{6.10}$$

 $<sup>^2\</sup>mathrm{Ratio}$  between gas and particle absorption coefficient is here higher, between 0.6 and 0.8 near the walls.

 $<sup>^{3}</sup>$ The model does not distinguish between coal and ash particles with respect to optical properties.

where  $d_{part,n}$  is the particle diameter of particle n.

In order to improve the agreement between measured and simulated radiative heat fluxes, particle emissivity is set to 0.7 in case 5 and case 6. This univariant sensitivity analysis covers a plausible range of uncertainty regarding coal and ash particle emissivity, cf. reference [87]. The resulting ratio between gas and particle absorption coefficient is shown in figure 6.11. Gas absorption coefficients in test cases 5 and 6 are essentially the same as those in cases 3 and 4, respectively, and therefore not shown here. Figure 6.11 shows that the ratio between gas and particle absorption coefficient is slightly higher in case 5 (< 0.333) compared to case 3 (< 0.2). In case 6, the ratio remains below 0.067 just like in case 4. Therefore, decreasing particle emissivity from 0.9 to 0.7 does not lead to significant lower particle absorption coefficients.

As a result, particle absorption coefficient and, thus, incident radiative heat fluxes remain high. Values of incident radiative heat flux in case 5 and 6 are shown in figure 6.12. Although particle emissivity is lower, predicted radiative heat fluxes exceed measured values as can be seen from figure 6.9.

In case 7, see table 6.1, reflectivity  $\rho_w$  of the furnace walls is therefore assumed to be 0.15. Results on gas temperature and radiative heat flux can be seen from figure 6.13. Gas temperature prediction on furnace level 1 and 6 is similar as in cases 3, 4 and 5, see also figures 6.7 and 6.8. Heat flux results, however, slightly improve in comparison with cases 3, 4 and 5, see also figure 6.9. Figure 6.14 shows plots of gas temperature at the furnace center and radiative heat flux at the furnace front wall according to the assumptions made in case 7.



(a) Cosine, level 1

(b) Case 3, level 1

Figure 6.7: Measured (symbols) and modeled (lines) gas temperatures on furnace level 1 ( $z_d = 1870$  mm): (a) cosine approximation according to section 4.2.1, (b) case 3, (c) case 4 and (d) case 5, see table 6.1


(a) Cosine, level 6

(b) Case 3, level 6

Figure 6.8: Measured (symbols) and modeled (lines) gas temperatures on furnace level 6 ( $z_d = 9140$  mm): (a) cosine approximation according to section 4.2.1, (b) case 3, (c) case 4 and (d) case 5, see table 6.1



Figure 6.9: Measured (symbols) and modeled radiative heat flux at the center of the furnace front wall (cases 3, 4 and 5 in table 6.1)



Figure 6.10: Ratio between gas and particle absorption coefficient at the furnace center cross–section (case 3 and case 4 in table 6.1)



Figure 6.11: Ratio between gas and particle absorption coefficient at the furnace center cross–section (case 5 and case 6 in table 6.1)



Figure 6.12: Incident radiative heat flux  $[kW/m^2]$  at the furnace front wall (case 5 and case 6 in table 6.1)



Figure 6.13: Case 7 in table 6.1: (a) measured and calculated gas temperature on furnace level 1 ( $z_d = 1870 \text{ mm}$ ) and 6 ( $z_d = 9140 \text{ mm}$ ) against furnace width in y-direction; (a) measured and calculated radiative heat fluxes at the center of the furnace front wall against distance from the furnace top  $z_d$ 



Figure 6.14: For case 7 in table 6.1: (a) gas temperature [°C] at the center crosssection of the furnace; (b) incident radiative heat flux [kW/m<sup>2</sup>] at the furnace front wall

### 7 Summary

Since exact solutions of the radiative transfer equation require high computational effort, simplified approaches are applied in engineering calculations. Radiative heat transfer is considered as being gray which means that the radiative transfer equation is solved on the basis of spectrally averaged radiative properties of gaseous and solid flue gas components instead of multiple solutions for a number of wavelengths or wavelength intervals. Moreover, absorption and emission coefficient are assumed to be equal. By this, computational effort of heat transfer simulation is drastically reduced. Calculation of heat transfer by radiation requires suitable simplified optical property models for radiating flue gas species, the most important being carbon dioxide and water vapor. These correlations describe the emissivity of gas mixtures as intensive quantity depending on gas composition and temperature. Solution of the differential, gray radiative transfer equation requires an extensive absorption coefficient. The absorption coefficient describes attenuation of heat radiation by gas per unit optical path length. Although the gas absorption coefficient is a strong function of wavelength, there are possibilities to calculate spectrally averaged absorption coefficients. In case of air-blown combustion, it is common to apply an "effective absorption coefficient" derived from Lambert–Beer's law and depending on gray emissivity and optical path length.

In case of oxy-fuel combustion, both carbon dioxide and water vapor concentration in the flue gas are higher than in that of air-blown combustion systems. Therefore, it is necessary to use gas emissivity correlations suited for high concentrations of these species. The widely applied weighted sum of gray gases correlation of Smith et al. [122] is only valid at certain mole fraction ratios of water vapor and carbon dioxide. In oxy-fuel combustion systems, this ratio depends on several factors such as the type of flue gas recirculation and fuel properties. A gas emissivity correlation based on a weighted sum of four gray gases and one transparent gas and suitable for high concentrations of radiating flue gas species allows for different mole fraction ratios and is applied in this work. Radiative wall heat fluxes in a 30 MW<sub>th</sub> oxy–fuel boiler are calculated with the help of a non–iterative Monte Carlo simulation. The calculation is based on fixed temperature profiles derived from measured values in order to check the quality of the applied emissivity correlation by comparing measured and calculated incident radiative heat fluxes. Results show that the gas absorption coefficient is low compared to the particle absorption coefficient. Increase of radiating gas species concentration due to oxy–fuel combustion has only little effect on radiative heat flux. Gas radiation contributes to a share of approximately 30 % to the total radiative heat flux. If gas temperatures and particle concentration are assumed to be equal in air–blown and oxy–fuel combustion, it is shown that the incident radiative heat flux at the center of the furnace front wall is on average 7.3 % higher in oxy–fuel combustion due to elevated concentrations of radiating gas species.

Calculation of the extensive spectrally averaged absorption coefficient as either effective or Patch absorption coefficient [95] is indentified to be more influential than the applied emissivity model. Calculation of an effective absorption coefficient from emissivity data based on Smith's model [122] leads to strong overestimation of radiative heat flux at elevated concentrations of carbon dioxide and water vapor. By contrast, application of Patch's average absorption coefficient yields radiative heat flux results which are in good agreement with measured values.

Incident radiative heat fluxes increase with increasing gas temperature. Increase in gas temperature leads to much stronger increase in radiative heat flux than increase in gas absorption coefficient. Therefore, higher gas absorption coefficients due to elevated concentrations of carbon dioxide and water vapor can be compensated by slight changes in gas temperature.

In practice, this can be done by changing the amount of recirculated flue gas. An oxidant mixture of flue gas and oxygen with an oxygen concentration of 28 % is identified to give similar radiative heat transfer to the furnace walls as air-blown combustion.

There is no evidence that quality of heat flux predictions in oxy-fuel furnaces by using gray models is lower with respect to such modeling of air-blown combustion systems. Moreover, application of Smith's model [122] which is intended for combustion with air shows only minor errors if the spectrally averaged absorption coefficient is derived as Patch absorption coefficient. Application of the effective absorption coefficient, however, overpredicts incident radiative heat fluxes at the furnace walls at relevant optical path lengths with regard to both tested emissivity correlations and oxy-fuel atmosphere. Results from iterative solution of the radiative transfer equation provide information on the effect of radiative properties of flue gas on both temperature and radiative heat flux. The non-iterative solution by Monte Carlo ray tracing in this work solves the radiative transfer equation based on invariant temperatures. However, temperatures are usually unknown and have to be calculated from energy balances. In order to evaluate the influence of gas optical properties in iterative solutions, an emissivity correlation suited for elevated concentrations of carbon dioxide and water vapor is implemented in a computational fluid dynamics code which is then used to simulate radiative heat transfer in the regarded oxy-fuel furnace.

If particle radiation is neglected, simulation based on effective absorption coefficient yields lower gas temperatures and higher incident radiative wall fluxes compared to modeling based on Patch absorption coefficient. This is due to higher emittance resulting in lower internal energy and, hence, temperatures. This is not compensated by higher gas absorptivity. Wall heat fluxes from effective absorption coefficient based calculation are approximately 50 % higher than those resulting from Patch based simulation. These results are only valid if radiation by soot, coal and ash particles is neglected.

If particle radiation is included in the simulation and particle emissivity is assumed to be 0.9, temperatures decrease and wall fluxes increase compared to pure gas atmosphere. The reason is higher emittance of the gas-particle-mixture. Modeling results show that flue gas composition becomes less relevant if particles are present in the flue gas. Temperature and heat flux results of Patch and effective absorption coefficient based simulations become very similar. Particle absorption coefficient is a multiple of gas absorption coefficient in both cases.

Comparison of modeled and measured values shows overestimation of incident radiative heat flux if particle emissivity and wall reflectivity are assumed to be 0.9 and 0.3, respectively. The particle absorption coefficient is overrated compared to published particle absorption coefficients [72]. Reduction of particle emissivity to 0.7 and assuming wall reflectivity to be 0.15 improves the agreement with measured values.

In general, the simulation shows that gas absorption coefficient not only influences wall heat fluxes but also gas temperature and, hence, all temperature dependent parameters. However, the influence of gas absorption coefficient on gas temperature is rather small since emission of radiative heat is only a linear function of absorption coefficient but a function of the fourth power of temperature. General shortcomings of simplifications like mean optical path length, diffuse reflection, isotropic scattering, spectrally averaged (gray) absorption coefficients and equality of gray emission and absorption coefficient only allow basic heat transfer calculations and estimation of design criteria of steam generators such as furnace exit gas temperature or wall heat fluxes. If more detailed results are desired, the computational effort increases and simplified, gray modeling might not be sufficient. The suitability of simplified methods for radiative transfer simulation has to be evaluated regarding computation time and desired results. There are more sophisticated gas emissivity models available (e.g., the wide band correlated–k model [128], the absorption distribution function with fictitious gases model [98, 99]) with acceptable increase in required computation time compared to weighted sum of gray gases correlations.

However, the weighted sum of gray gases model remains widely applied in engineering calculations. It is proven in this work that application of a modified weighted sum of gray gases model leads to satisfactory results in the simulation of an industrial oxy-fuel furnace. Quality of the results is similar to that of air-blown combustion simulation. It can be concluded that several gas emissivity correlations are available providing sufficiently accurate results. More than accurate calculation of gas emissivity, the calculation of spectrally averaged absorption coefficients is crucial in order to yield acceptable results from gray modeling. Patch's absorption coefficient or any other suitable spectrally averaged absorption coefficient should be applied.

# A WSGG Model Parameters

## A.1 Polynomial Coefficients for Weighting Factor Calculation

Table A.1: Coefficients for weighting factor calculation as function of  $H_2O/CO_2$  mole fraction ratio

u	v	$c_{\epsilon,u,v,1}$	$c_{\epsilon,u,v,2}$	$c_{\epsilon,u,v,3}$	$c_{\epsilon,u,v,4}$
			$1/18 \leq \Psi < 3$	3/13	
1	1	$1.69456 \cdot 10^{+00}$	$-1.31000 \cdot 10^{+00}$	$-8.95328 \cdot 10^{-02}$	$6.34644 \cdot 10^{-01}$
2	1	$-2.77827 \cdot 10^{+00}$	$2.08303 \cdot 10^{+00}$	$-1.68456 \cdot 10^{-01}$	$3.14235 \cdot 10^{-01}$
3	1	$-1.10817 \cdot 10^{-01}$	$1.32398 \cdot 10^{-02}$	$2.00309 \cdot 10^{-02}$	$1.02102 \cdot 10^{-01}$
4	1	$-1.28290 \cdot 10^{+00}$	$8.46219 \cdot 10^{-01}$	$1.93415 \cdot 10^{-01}$	$-3.58416 \cdot 10^{-03}$
1	2	$-2.98882 \cdot 10^{-04}$	$3.05214 \cdot 10^{-04}$	$4.21836 \cdot 10^{-05}$	$-8.07175 \cdot 10^{-04}$
2	2	$3.31170 \cdot 10^{-03}$	$-2.47367 \cdot 10^{-03}$	$2.28752 \cdot 10^{-04}$	$-2.39068 \cdot 10^{-05}$
3	2	$-3.08130 \cdot 10^{-05}$	$1.78431 \cdot 10^{-05}$	$3.65210 \cdot 10^{-05}$	$9.48794 \cdot 10^{-05}$
4	2	$2.10199 \cdot 10^{-03}$	$-1.67361 \cdot 10^{-03}$	$1.94370 \cdot 10^{-04}$	$1.06024 \cdot 10^{-04}$
1	3	$3.23002 \cdot 10^{-07}$	$-5.24798 \cdot 10^{-07}$	$3.20655 \cdot 10^{-07}$	$5.28509 \cdot 10^{-07}$
<b>2</b>	3	$-1.19488 \cdot 10^{-06}$	$9.44801 \cdot 10^{-07}$	$-1.00100 \cdot 10^{-07}$	$-4.91082 \cdot 10^{-08}$
3	3	$-1.40438 \cdot 10^{-06}$	$1.02776 \cdot 10^{-06}$	$-5.51703 \cdot 10^{-08}$	$-9.37148 \cdot 10^{-08}$
4	3	$-8.65291 \cdot 10^{-07}$	$8.48984 \cdot 10^{-07}$	$-3.11117 \cdot 10^{-07}$	$-8.55165 \cdot 10^{-08}$
1	4	$-1.82115 \cdot 10^{-10}$	$2.24905 \cdot 10^{-10}$	$-1.13832 \cdot 10^{-10}$	$-1.09961 \cdot 10^{-10}$
<b>2</b>	4	$1.32229 \cdot 10^{-10}$	$-1.29807 \cdot 10^{-10}$	$2.46630 \cdot 10^{-11}$	$1.23543 \cdot 10^{-11}$
3	4	$5.08207 \cdot 10^{-10}$	$-3.52602 \cdot 10^{-10}$	$1.79773 \cdot 10^{-12}$	$1.80559 \cdot 10^{-11}$
4	4	$1.06847 \cdot 10^{-10}$	$-1.43968 \cdot 10^{-10}$	$8.86638 \cdot 10^{-11}$	$1.80801 \cdot 10^{-11}$
			$3/13 \leq \Psi < 1$	1/27	
1	1	$1.69456 \cdot 10^{+00}$	$-4.15682 \cdot 10^{-01}$	$-3.93116 \cdot 10^{-01}$	$5.87578 \cdot 10^{-01}$
$^{2}$	1	$-2.77827 \cdot 10^{+00}$	$6.16768 \cdot 10^{-01}$	$3.06492 \cdot 10^{-01}$	$3.33939 \cdot 10^{-01}$
3	1	$-1.10817 \cdot 10^{-01}$	$-4.52453 \cdot 10^{-02}$	$1.44005 \cdot 10^{-02}$	$1.05432 \cdot 10^{-01}$
4	1	$-1.28290 \cdot 10^{+00}$	$1.69153 \cdot 10^{-01}$	$3.72040 \cdot 10^{-01}$	$4.96456 \cdot 10^{-02}$
1	<b>2</b>	$-2.98882 \cdot 10^{-04}$	$1.47476 \cdot 10^{-04}$	$1.21821 \cdot 10^{-04}$	$-7.91935 \cdot 10^{-04}$
<b>2</b>	<b>2</b>	$3.31170 \cdot 10^{-03}$	$-7.25891 \cdot 10^{-04}$	$-3.34116 \cdot 10^{-04}$	$-4.21897 \cdot 10^{-05}$
3	<b>2</b>	$-3.08130 \cdot 10^{-05}$	$1.58126 \cdot 10^{-06}$	$3.99381 \cdot 10^{-05}$	$1.01688 \cdot 10^{-04}$
4	<b>2</b>	$2.10199 \cdot 10^{-03}$	$-5.64271 \cdot 10^{-04}$	$-1.99319 \cdot 10^{-04}$	$9.98669 \cdot 10^{-05}$
1	3	$3.23002 \cdot 10^{-07}$	$-3.54330 \cdot 10^{-07}$	$1.65999 \cdot 10^{-07}$	$5.70436 \cdot 10^{-07}$
<b>2</b>	3	$-1.19488 \cdot 10^{-06}$	$3.14189 \cdot 10^{-07}$	$1.21380 \cdot 10^{-07}$	$-4.39837 \cdot 10^{-08}$
3	3	$-1.40438 \cdot 10^{-06}$	$2.86590 \cdot 10^{-07}$	$1.76051 \cdot 10^{-07}$	$-7.92591 \cdot 10^{-08}$
4	3	$-8.65291 \cdot 10^{-07}$	$3.92317 \cdot 10^{-07}$	$-9.27479 \cdot 10^{-08}$	$-1.18685 \cdot 10^{-07}$
1	4	$-1.82115 \cdot 10^{-10}$	$1.28792 \cdot 10^{-10}$	$-5.16098 \cdot 10^{-11}$	$-1.24017 \cdot 10^{-10}$
<b>2</b>	4	$1.32229 \cdot 10^{-10}$	$-6.00214 \cdot 10^{-11}$	$-8.73160 \cdot 10^{-12}$	$1.33957 \cdot 10^{-11}$
3	4	$5.08207 \cdot 10^{-10}$	$-8.43907 \cdot 10^{-11}$	$-7.50781 \cdot 10^{-11}$	$1.02267 \cdot 10^{-11}$

u	v	$c_{\epsilon,u,v,1}$	$c_{\epsilon,u,v,2}$	$c_{\epsilon,u,v,3}$	$c_{\epsilon,u,v,4}$
4	4	$1.06847 \cdot 10^{-10}$	$-8.75786 \cdot 10^{-11}$	$4.79299 \cdot 10^{-11}$	$2.98040 \cdot 10^{-11}$
			$11/27 \leq \Psi < 4$	41/54	
1	1	$-1.68245 \cdot 10^{-01}$	$4.78690 \cdot 10^{-01}$	$-3.82031 \cdot 10^{-01}$	$5.14778 \cdot 10^{-01}$
2	1	$8.48056 \cdot 10^{-01}$	$-8.49576 \cdot 10^{-01}$	$2.65534 \cdot 10^{-01}$	$3.91822 \cdot 10^{-01}$
3	1	$1.44574 \cdot 10^{-01}$	$-1.03733 \cdot 10^{-01}$	$-1.18093 \cdot 10^{-02}$	$1.05962 \cdot 10^{-01}$
4	1	$3.34558 \cdot 10^{-01}$	$-5.07950 \cdot 10^{-01}$	$3.12435 \cdot 10^{-01}$	$1.13348 \cdot 10^{-01}$
1	2	$-1.41140 \cdot 10^{-04}$	$-1.02711 \cdot 10^{-05}$	$1.45959 \cdot 10^{-04}$	$-7.67566 \cdot 10^{-04}$
2	<b>2</b>	$-1.13496 \cdot 10^{-03}$	$1.02199 \cdot 10^{-03}$	$-2.82022 \cdot 10^{-04}$	$-1.05405 \cdot 10^{-04}$
3	<b>2</b>	$1.03354 \cdot 10^{-04}$	$-1.46815 \cdot 10^{-05}$	$3.76334 \cdot 10^{-05}$	$1.08596 \cdot 10^{-04}$
4	2	$-5.52568 \cdot 10^{-04}$	$5.45140 \cdot 10^{-04}$	$-2.02685 \cdot 10^{-04}$	$5.87815 \cdot 10^{-05}$
1	3	$2.04614 \cdot 10^{-07}$	$-1.83853 \cdot 10^{-07}$	$7.13165 \cdot 10^{-08}$	$5.90432 \cdot 10^{-07}$
2	3	$2.78325 \cdot 10^{-07}$	$-3.16458 \cdot 10^{-07}$	$1.20981 \cdot 10^{-07}$	$-1.94110 \cdot 10^{-08}$
3	3	$4.11464 \cdot 10^{-07}$	$-4.54630 \cdot 10^{-07}$	$1.46488 \cdot 10^{-07}$	$-4.70632 \cdot 10^{-08}$
4	3	$1.28955 \cdot 10^{-07}$	$-6.43746 \cdot 10^{-08}$	$-3.50529 \cdot 10^{-08}$	$-1.27571 \cdot 10^{-07}$
1	4	$-4.01049 \cdot 10^{-11}$	$3.26734 \cdot 10^{-11}$	$-2.32032 \cdot 10^{-11}$	$-1.30103 \cdot 10^{-10}$
2	4	$2.94024 \cdot 10^{-11}$	$9.76820 \cdot 10^{-12}$	$-1.75726 \cdot 10^{-11}$	$1.07218 \cdot 10^{-11}$
3	4	$-1.94616 \cdot 10^{-10}$	$1.83836 \cdot 10^{-10}$	$-5.75826 \cdot 10^{-11}$	$-2.82641 \cdot 10^{-12}$
4	4	$1.97989 \cdot 10^{-11}$	$-3.11854 \cdot 10^{-11}$	$2.70358 \cdot 10^{-11}$	$3.61075 \cdot 10^{-11}$
			$41/54 \le \Psi < 1$	0/09	
1	1	$-2.74585\dot{1}0^{-01}$	$3.01098\dot{1}0^{-01}$	$-1.07661\dot{1}0^{-01}$	$4.32293\dot{1}0^{-01}$
2	1	$-9.4673510^{-02}$	$4.5589910^{-02}$	$-1.73478\dot{1}0^{-02}$	$4.17014\dot{1}0^{-01}$
3	1	$-2.2532210^{-02}$	$4.88716\dot{1}0^{-02}$	$-3.1112510^{-02}$	$9.5262810^{-02}$
4	1	$9.30965\dot{1}0^{-02}$	$-1.5480710^{-01}$	$7.9244410^{-02}$	$1.74968\dot{1}0^{-01}$
1	<b>2</b>	$1.51410\dot{1}0^{-04}$	$-1.59251\dot{1}0^{-04}$	$8.63128\dot{1}0^{-05}$	$-7.2363010^{-04}$
2	2	$2.71400\dot{1}0^{-04}$	$-1.7602310^{-04}$	$1.56322\dot{1}0^{-05}$	$-1.27551\dot{1}0^{-04}$
3	2	$-1.26184\dot{1}0^{-04}$	$9.44138\dot{1}0^{-05}$	$6.56872\dot{1}0^{-05}$	$1.24521\dot{1}0^{-04}$
4	2	$5.15965\dot{1}0^{-05}$	$-3.81235\dot{1}0^{-05}$	$-2.42914\dot{1}0^{-05}$	$3.08852\dot{1}0^{-05}$
1	3	$-6.51422\dot{1}0^{-08}$	$3.21275\dot{1}0^{-08}$	$1.79318\dot{1}0^{-08}$	$6.01677\dot{1}0^{-07}$
2	3	$-1.64356\dot{1}0^{-08}$	$-2.26716\dot{1}0^{-08}$	$1.65889\dot{1}0^{-09}$	$-3.89723\dot{1}0^{-09}$
3	3	$-6.8682710^{-09}$	$-2.0309210^{-08}$	$-2.0619210^{-08}$	$-3.38810\dot{1}0^{-08}$
4	3	$-3.6303610^{-08}$	$7.17442\dot{1}0^{-08}$	$-3.2459810^{-08}$	$-1.42257\dot{1}0^{-07}$
1	4	$2.01662\dot{1}0^{-11}$	$-9.65937\dot{1}0^{-12}$	$-1.51057\dot{1}0^{-11}$	$-1.35969\dot{1}0^{-10}$
$^{2}$	4	$-3.5480410^{-11}$	$4.08040\dot{1}0^{-11}$	$2.21180\dot{1}0^{-13}$	$7.02895\dot{1}0^{-12}$
3	4	$3.4688310^{-11}$	$-2.15913\dot{1}0^{-11}$	$-4.96811\dot{1}0^{-13}$	$-8.80544\dot{1}0^{-12}$
4	4	$-2.7826910^{-12}$	$-1.0286610^{-11}$	$1.24438\dot{1}0^{-11}$	$4.26217\dot{1}0^{-11}$

Table A.1: Coefficients for weighting factor calculation as function of  $H_2O/CO_2$  mole fraction ratio (continued)

u	v	$c_{\epsilon,u,v,1}$	$c_{\epsilon,u,v,2}$	$c_{\epsilon,u,v,3}$	$c_{\epsilon,u,v,4}$		
	$10/9 \le \Psi < 79/54$						
1	1	$-1.22306 \cdot 10^{-02}$	$1.12603 \cdot 10^{-02}$	$2.24164 \cdot 10^{-03}$	$4.19727 \cdot 10^{-01}$		
2	1	$1.20688 \cdot 10^{-01}$	$-5.43427 \cdot 10^{-02}$	$-2.04275 \cdot 10^{-02}$	$4.12430 \cdot 10^{-01}$		
3	1	$2.26035 \cdot 10^{-03}$	$2.50877 \cdot 10^{-02}$	$-5.08995 \cdot 10^{-03}$	$8.93846 \cdot 10^{-02}$		
4	1	$5.91969 \cdot 10^{-02}$	$-5.65391 \cdot 10^{-02}$	$4.88219 \cdot 10^{-03}$	$1.87740 \cdot 10^{-01}$		
1	2	$-4.57155 \cdot 10^{-05}$	$5.69986 \cdot 10^{-07}$	$3.04806 \cdot 10^{-05}$	$-7.06381 \cdot 10^{-04}$		
2	2	$-1.89411 \cdot 10^{-04}$	$1.10453 \cdot 10^{-04}$	$-7.43846 \cdot 10^{-06}$	$-1.32020 \cdot 10^{-04}$		
3	<b>2</b>	$-6.16263 \cdot 10^{-05}$	$-3.87805 \cdot 10^{-05}$	$8.52618 \cdot 10^{-05}$	$1.53825 \cdot 10^{-04}$		
4	<b>2</b>	$3.85531 \cdot 10^{-05}$	$1.63392 \cdot 10^{-05}$	$-3.19562 \cdot 10^{-05}$	$1.98661 \cdot 10^{-05}$		
1	3	$3.60731 \cdot 10^{-08}$	$-3.66333 \cdot 10^{-08}$	$1.63465 \cdot 10^{-08}$	$6.09126 \cdot 10^{-07}$		
2	3	$9.80167 \cdot 10^{-08}$	$-4.00203 \cdot 10^{-08}$	$-2.03992 \cdot 10^{-08}$	$-6.83617 \cdot 10^{-09}$		
3	3	$1.16521 \cdot 10^{-07}$	$-2.75590 \cdot 10^{-08}$	$-3.74616 \cdot 10^{-08}$	$-4.39493 \cdot 10^{-08}$		
4	3	$-6.63634 \cdot 10^{-08}$	$3.34239 \cdot 10^{-08}$	$4.54351 \cdot 10^{-09}$	$-1.46377 \cdot 10^{-07}$		
1	4	$1.39691 \cdot 10^{-12}$	$1.16270 \cdot 10^{-11}$	$-1.44133 \cdot 10^{-11}$	$-1.41601 \cdot 10^{-10}$		
$^{2}$	4	$-2.33522 \cdot 10^{-11}$	$3.35264 \cdot 10^{-12}$	$1.57576 \cdot 10^{-11}$	$1.06127 \cdot 10^{-11}$		
3	4	$-2.97895 \cdot 10^{-11}$	$1.50239 \cdot 10^{-11}$	$-2.80756 \cdot 10^{-12}$	$-1.01422 \cdot 10^{-11}$		
4	4	$1.51786 \cdot 10^{-11}$	$-1.32239 \cdot 10^{-11}$	$4.17168 \cdot 10^{-12}$	$4.56054 \cdot 10^{-11}$		
			$79/54 \le \Psi < 4$	19/27			
1	1	$-4.03408i0^{-03}$	$-1.64968i0^{-03}$	$5.62317\dot{1}0^{-03}$	$4.21377\dot{1}0^{-01}$		
2	1	$-7.29388i0^{-02}$	$7.30504\dot{1}0^{-02}$	$-1.38452\dot{1}0^{-02}$	$4.03773\dot{1}0^{-01}$		
3	1	$-4.82428i0^{-02}$	$2.74736\dot{1}0^{-02}$	$1.34037\dot{1}0^{-02}$	$9.07980\dot{1}0^{-02}$		
4	1	$7.49295\dot{10}^{-03}$	$5.94621\dot{1}0^{-03}$	$-1.2918910^{-02}$	$1.85037\dot{1}0^{-01}$		
1	2	$4.10158\dot{10}^{-05}$	$-4.7685110^{-05}$	$1.39032\dot{1}0^{-05}$	$-6.9757710^{-04}$		
$^{2}$	<b>2</b>	$1.07824\dot{1}0^{-04}$	$-8.94793i0^{-05}$	$-5.86758\dot{1}0^{-08}$	$-1.29214\dot{1}0^{-04}$		
3	<b>2</b>	$8.60623 \dot{10}^{-05}$	$-1.03830\dot{1}0^{-04}$	$3.50842\dot{1}0^{-05}$	$1.76339\dot{1}0^{-04}$		
4	2	$-6.27312\dot{10}^{-05}$	$5.7033910^{-05}$	$-6.1398610^{-06}$	$1.23243\dot{1}0^{-05}$		
1	3	$-6.0398710^{-09}$	$1.44365\dot{1}0^{-09}$	$3.9650310^{-09}$	$6.11914\dot{1}0^{-07}$		
2	3	$-6.3397410^{-08}$	$6.34412\dot{1}0^{-08}$	$-1.2158610^{-08}$	$-1.4698610^{-08}$		
3	3	$-1.01457\dot{10}^{-07}$	$9.54356\dot{1}0^{-08}$	$-1.35792\dot{1}0^{-08}$	$-5.5466510^{-08}$		
4	3	$4.18719\dot{1}0^{-08}$	$-3.6626010^{-08}$	$3.4168610^{-09}$	$-1.4353110^{-07}$		
1	4	$-8.66375\dot{1}0^{-12}$	$1.31015\dot{1}0^{-11}$	$-5.71259\dot{1}0^{-12}$	$-1.45172\dot{1}0^{-10}$		
<b>2</b>	4	$1.44330\dot{1}0^{-11}$	$-2.12968i0^{-11}$	$9.44403\dot{1}0^{-12}$	$1.55549\dot{1}0^{-11}$		
3	4	$2.43172\dot{1}0^{-11}$	$-1.64204\dot{1}0^{-11}$	$-3.29891\dot{1}0^{-12}$	$-1.05677\dot{1}0^{-11}$		
4	4	$-4.45940\dot{1}0^{-12}$	$2.79792\dot{1}0^{-12}$	$5.03303\dot{1}0^{-13}$	$4.60973\dot{1}0^{-11}$		
			$49/27 \leq \Psi <$	13/6			
1	1	$-4.03408 \cdot 10^{-03}$	$-5.90786 \cdot 10^{-03}$	$2.96405 \cdot 10^{-03}$	$4.22976 \cdot 10^{-01}$		

Table A.1: Coefficients for weighting factor calculation as function of  $H_2O/CO_2$  mole fraction ratio (continued)

u	v	$c_{\epsilon,u,v,1}$	$c_{\epsilon,u,v,2}$	$c_{\epsilon,u,v,3}$	$c_{\epsilon,u,v,4}$
2	1	$-7.29388 \cdot 10^{-02}$	$-3.94022 \cdot 10^{-03}$	$1.04711 \cdot 10^{-02}$	$4.04768 \cdot 10^{-01}$
3	1	$-4.82428 \cdot 10^{-02}$	$-2.34490 \cdot 10^{-02}$	$1.48197 \cdot 10^{-02}$	$9.68139 \cdot 10^{-02}$
4	1	$7.49295 \cdot 10^{-03}$	$1.38554 \cdot 10^{-02}$	$-5.95172 \cdot 10^{-03}$	$1.81554 \cdot 10^{-01}$
1	<b>2</b>	$4.10158 \cdot 10^{-05}$	$-4.39088 \cdot 10^{-06}$	$-4.41973 \cdot 10^{-06}$	$-6.96802 \cdot 10^{-04}$
2	2	$1.07824 \cdot 10^{-04}$	$2.43347 \cdot 10^{-05}$	$-2.29798 \cdot 10^{-05}$	$-1.35615 \cdot 10^{-04}$
3	2	$8.60623 \cdot 10^{-05}$	$-1.29871 \cdot 10^{-05}$	$-6.01790 \cdot 10^{-06}$	$1.79579 \cdot 10^{-04}$
4	<b>2</b>	$-6.27312 \cdot 10^{-05}$	$-9.18198 \cdot 10^{-06}$	$1.06968 \cdot 10^{-05}$	$1.44923 \cdot 10^{-05}$
1	3	$-6.03987 \cdot 10^{-09}$	$-4.93173 \cdot 10^{-09}$	$2.73775 \cdot 10^{-09}$	$6.13224 \cdot 10^{-07}$
<b>2</b>	3	$-6.33974 \cdot 10^{-08}$	$-3.47791 \cdot 10^{-09}$	$8.93948 \cdot 10^{-09}$	$-1.38842 \cdot 10^{-08}$
3	3	$-1.01457 \cdot 10^{-07}$	$-1.16574 \cdot 10^{-08}$	$1.58980 \cdot 10^{-08}$	$-5.28489 \cdot 10^{-08}$
4	3	$4.18719 \cdot 10^{-08}$	$7.57197 \cdot 10^{-09}$	$-6.80580 \cdot 10^{-09}$	$-1.45039 \cdot 10^{-07}$
1	4	$-8.66375 \cdot 10^{-12}$	$3.95656 \cdot 10^{-12}$	$2.89320 \cdot 10^{-13}$	$-1.45937 \cdot 10^{-10}$
2	4	$1.44330 \cdot 10^{-11}$	$-6.06204 \cdot 10^{-12}$	$-1.82176 \cdot 10^{-13}$	$1.68700 \cdot 10^{-11}$
3	4	$2.43172 \cdot 10^{-11}$	$9.24763 \cdot 10^{-12}$	$-5.82266 \cdot 10^{-12}$	$-1.27020 \cdot 10^{-11}$
4	4	$-4.45940 \cdot 10^{-12}$	$-1.90920 \cdot 10^{-12}$	$8.16001 \cdot 10^{-13}$	$4.64265 \cdot 10^{-11}$

Table A.1: Coefficients for weighting factor calculation as function of  $H_2O/CO_2$  mole fraction ratio (continued)

#### A.2 Polynomial Coefficients for Absorption Coefficient Calculation

Table	A.2:	$\operatorname{Coefficients}$	$\mathbf{for}$	absorption	$\operatorname{coefficient}$	calculation	$^{\mathrm{as}}$	function	$\mathbf{of}$	$H_2O$	$/CO_2$
		mole fractio	rat	io							

u	$d_{u,1}$	$d_{u,2}$	$d_{u,3}$	$d_{u,4}$
		$1/18 \leq \Psi <$	< 3/13	
$egin{array}{c} 1 \\ 2 \\ 3 \\ 4 \end{array}$	$\begin{array}{c} 1.30527 \cdot 10^{+00} \\ 1.36017 \cdot 10^{+01} \\ 3.59814 \cdot 10^{+01} \\ -7.47192 \cdot 10^{+03} \end{array}$	$\begin{array}{l} -1.23685 \cdot 10^{+00} \\ -1.30504 \cdot 10^{+01} \\ 6.77250 \cdot 10^{-01} \\ 8.52155 \cdot 10^{+03} \end{array}$	$\begin{array}{l} 3.23182 \cdot 10^{-01} \\ 3.69169 \cdot 10^{+00} \\ -2.51028 \cdot 10^{+01} \\ -3.49644 \cdot 10^{+03} \end{array}$	$\begin{array}{c} 4.81157 \cdot 10^{-02} \\ 5.41446 \cdot 10^{-01} \\ 1.39192 \cdot 10^{+01} \\ 6.19460 \cdot 10^{+02} \end{array}$
		$3/13 \leq \Psi <$	11/27	
1	$1.30527 \cdot 10^{+00}$	$-5.47984 \cdot 10^{-01}$	$9.19266 \cdot 10^{-03}$	$7.37982 \cdot 10^{-02}$

u	$d_{u,1}$	$d_{u,2}$	$d_{u,3}$	$d_{u,4}$
2	$1.36017 \cdot 10^{+01}$	$-5.87196 \cdot 10^{+00}$	$3.62873 \cdot 10^{-01}$	$8.61060 \cdot 10^{-01}$
3	$3.59814 \cdot 10^{+01}$	$1.96668 \cdot 10^{+01}$	$-2.15239 \cdot 10^{+01}$	$9.71999 \cdot 10^{+00}$
4	$-7.47192 \cdot 10^{+03}$	$4.57817 \cdot 10^{+03}$	$-1.19194 \cdot 10^{+03}$	$2.27408 \cdot 10^{+02}$
		$11/27 \leq \Psi <$	< 41/54	
1	$-8.90850 \cdot 10^{-02}$	$1.40927 \cdot 10^{-01}$	$-6.24208 \cdot 10^{-02}$	$6.55622 \cdot 10^{-02}$
2	$-7.53832 \cdot 10^{-01}$	$1.30690 \cdot 10^{+00}$	$-4.40256 \cdot 10^{-01}$	$8.17220 \cdot 10^{-01}$
3	$-3.83890 \cdot 10^{+01}$	$3.86574 \cdot 10^{+01}$	$-1.12629 \cdot 10^{+01}$	$6.73793 \cdot 10^{+00}$
4	$-5.24828 \cdot 10^{+02}$	$6.34567 \cdot 10^{+02}$	$-2.74867 \cdot 10^{+02}$	$1.18724 \cdot 10^{+02}$
		$41/54 \le \Psi$	< 10/9	
1	$-4.16637 \cdot 10^{-02}$	$4.68938 \cdot 10^{-02}$	$3.66406 \cdot 10^{-03}$	$5.71656 \cdot 10^{-02}$
2	$-5.45696 \cdot 10^{-01}$	$5.11195 \cdot 10^{-01}$	$1.99441 \cdot 10^{-01}$	$7.91273 \cdot 10^{-01}$
3	$3.96916 \cdot 10^{+00}$	$-1.86408 \cdot 10^{+00}$	$1.68281 \cdot 10^{+00}$	$5.88863 \cdot 10^{+00}$
4	$-3.81133 \cdot 10^{+01}$	$8.05854 \cdot 10^{+01}$	$8.05854 \cdot 10^{+01} \qquad -2.32403 \cdot 10^{+01}$	
		$10/9 \le \Psi <$	79/54	
1	$-5.50775 \cdot 10^{-02}$	$2.91565 \cdot 10^{-03}$	$2.11895 \cdot 10^{-02}$	$6.24454 \cdot 10^{-02}$
2	$-2.42803 \cdot 10^{-01}$	$-6.48136 \cdot 10^{-02}$	$3.56500 \cdot 10^{-01}$	$9.00962 \cdot 10^{-01}$
3	$-4.98995 \cdot 10^{+00}$	$2.32556 \cdot 10^{+00}$	$1.84518 \cdot 10^{+00}$	$6.42285 \cdot 10^{+00}$
4	$-7.20322 \cdot 10^{+01}$	$4.03548 \cdot 10^{+01}$	$1.93124 \cdot 10^{+01}$	$7.78489 \cdot 10^{+01}$
		$79/54 \le \Psi <$	< 49/27	
1	$8.11389 \cdot 10^{-02}$	$-5.52214 \cdot 10^{-02}$	$2.78572 \cdot 10^{-03}$	$6.78628 \cdot 10^{-02}$
2	$1.62629 \cdot 10^{-01}$	$-3.21104 \cdot 10^{-01}$	$2.20715 \cdot 10^{-01}$	$1.00779 \cdot 10^{+00}$
3	$2.57599 \cdot 10^{+00}$	$-2.94158 \cdot 10^{+00}$	$1.62843 \cdot 10^{+00}$	$7.14262 \cdot 10^{+00}$
4	$1.94693 \cdot 10^{+01}$	$-3.56787 \cdot 10^{+01}$	$2.09577 \cdot 10^{+01}$	$8.65022 \cdot 10^{+01}$
		$49/27 \le \Psi$	< 13/6	
1	$8.11389 \cdot 10^{-02}$	$3.04247 \cdot 10^{-02}$	$-5.93898 \cdot 10^{-03}$	$6.55409 \cdot 10^{-02}$
2	$1.62629 \cdot 10^{-01}$	$-1.49441 \cdot 10^{-01}$	$5.51540 \cdot 10^{-02}$	$1.05278 \cdot 10^{+00}$
3	$2.57599 \cdot 10^{+00}$	$-2.22498 \cdot 10^{-01}$	$5.15154 \cdot 10^{-01}$	$7.46363 \cdot 10^{+00}$
4	$1.94693 \cdot 10^{+01}$	$-1.51279 \cdot 10^{+01}$	$3.08136 \cdot 10^{+00}$	$9.03073 \cdot 10^{+01}$

Table A.2: Coefficients for absorption coefficient calculation as function of  $H_2O/CO_2$ mole fractio ratio (continued)

B Comparison of WSGGM and EWBM Emissivities



Figure B.1: Emissivity at different optical path lengths L as function of gas temperature  $T_g$  generated from EWBM (symbols) and WSGG4+1 (lines) for (a)  $\psi_{CO_2}=0.3$  and  $\psi_{H_2O}=0.65$  and (b)  $\psi_{CO_2}=0.9$  and  $\psi_{H_2O}=0.05$ .



Figure B.2: Emissivity at different optical path lengths L as function of gas temperature  $T_g$  generated from EWBM (symbols) and WSGG3+1 [122] (lines) for (a)  $\psi_{CO_2}=0.3$  and  $\psi_{H_2O}=0.65$  and (b)  $\psi_{CO_2}=0.9$  and  $\psi_{H_2O}=0.05$ .

## Bibliography

- Abraham, B. M.; Asbury, J. G.; Lynch, E. P.; Teotia, A. P. S.: Coal–Oxygen Process Provides CO<sub>2</sub> for Enhanced Oil Recovery; Oil and Gas Journal 80 (1982) 68-75.
- [2] Adams, B. R.; Smith, P. J.: Three-dimensional Discrete-ordinates Modeling of Radiative Transfer in a Geometrically Complex Furnace; Combustion Science and Technology 88 (1993) 293-308.
- [3] Al-Makhadmeh, L.; Maier, J.; Scheffknecht, G.: Coal Pyrolysis and Char Combustion under Oxy-Fuel Conditions; Proceedings of the 34th International Technical Conference on Coal Utilization and Fuel Systems (2009).
- [4] Andersson, K.; Johansson, R.; Hjärtstam, S.; Johnsson, F.; Leckner, B.: Radiation Intensity of Lignite–Fired Oxy–Fuel Flames; Experimental Thermal and Fluid Science 33 (2008) 67-76.
- [5] Andersson, K.; Johansson, R.; Johnsson, F.; Leckner, B.: Radiation Intensity of Propane–Fired Oxy–Fuel Flames: Implications for Soot Formation; Energy & Fuels 22 (2008) 1535-1541.
- [6] Andersson, K.; Johnsson, F.: Flame and Radiation characteristics of gas fired O<sub>2</sub>/CO<sub>2</sub> combustion; Fuel 86 (2007) 656-668.
- [7] Bartelds, H., Heap, M. P., Lowes, T. M.: Radiative Heat Transfer in Enclosures; IFRF Doc. no. G04/A/6 (1977).
- [8] Becher, V.; Goanta, A.; Spliethoff, H.: Validation of spectral gas radiation models under oxyfuel conditions — Part C: Validation of simplified models; International Journal of Greenhouse Gas Control 11 (2012) 34-51.
- [9] Bejan, A.; Kraus, A. D.; Heat Transfer Handbook; John Wiley & Sons, Inc., Hoboken, New Jersey (2003).

- [10] Brunn, O.: Modellierung des dreidimensionalen Strahlungswärmeaustauschs in Verbrennungsräumen mittels Monte Carlo Methode; Universität Karlsruhe, Falkutät für Chemieingenieurwesen und Verfahrenstechnik, Dissertation (2010).
- [11] Buckius, R. O.; Hwang, D. C.: Radiation Properties for Polydispersions: Application to Coal; ASME Journal of Heat Transfer 102 (1980) 99-103.
- [12] Buhre, B. J. P.; Elliot, L. K.; Sheng, C. D.; Gupta, R. P.; Wall, T. F.: Oxy– Fuel Combustion Technology for Coal–Fired Power Generation; Progress in Energy and Combustion Science 31 (2005) 283-307.
- [13] Burchhardt, U.; Radunsky, D.: Die Oxyfuel–Forschungsanlage von Vattenfall, 39. Kraftwerkstechnisches Kolloquium (2009).
- [14] Carvalho, M. G.; Coelho, P. J.; Moreira, A. L. N.; Silva, A. M. C.; Silva, T. F.: Comparison of Measurements and Predictions of Wall Heat Flux and Gas Composition in an Oil–Fired Utility Boiler; 25th Symposium (International) on Combustion (1994) 227-234.
- [15] Carvalho, M. G.; Farias, T. L.: Modelling of Heat Transfer in Radiating and Combustion Systems; Transaction of the IChemE 76, Part A (1998) 175-184.
- [16] Chang, H.; Charalampopoulos, T. T.: Determination of the wavelength dependence of refractive indices of flame soot; Proceedings Royal Society, Series A, 430 (1990) 577-591.
- [16] Chang, S. L.; Rhee, K. T.: Blackbody Radiation Functions; International Communications in Heat and Mass Transfer 11 (1984) 451-455.
- [17] Charalampopoulos, T. T.; Chang, H.: Effects of Soot Agglomeration on Radiative Transfer; J. Quant. Spectroc. Radiat. Transfer 46 (1991) 125-134.
- [18] Chedaille, J.; Braud, Y.: Measurements in Flames Volume 1; Edward Arnold Ltd., London (1972).
- [19] Chen, J.; Mann, A. P., Kent, J. H.: Computational Modelling of Pulverized Fuel Burnout in Tangentially Fired Furnaces; 24th Symposium (International) on Combustion (1992) 1381-1389.

- [20] Chui, E.; Raithby, G.: Computation of Radiant Heat Transfer on a Non– Orthogonal Mesh Using The Finite Volume Method; Numerical Heat Transfer, Part B, 23 (1993) 269-288.
- [21] Coppalle, A.; Vervish, P.: The Total Emissivities of High Temperature Flames; Combustion and Flame 49 (1986) 101-108.
- [22] Crosbie, A. L.; Farrell, J. B.: Exact Formulation of Multiple Scattering in a Three–Dimensional Cylindrical Geometry; Journal of Quantitative Spectroscopy and Radiative Transfer 31 (1984) 397-416.
- [23] Crosbie, A. L.; Schrenker, R. G.: Exact Expressions for Radiative Transfer in a Three–Dimensional Rectangular Geometry; Journal of Quantitative Spectroscopy and Radiative Transfer 28 (1982) 507-526.
- [24] Dalzell, W. H.; Sarofim, A. H.: Optical constants of soot and their application to heat-flux calculations; ASME Journal of Heat Transfer 91 (1969) 100-104.
- [25] Davidson, R. M.; Santos, S. O.: Oxyfuel combustion of pulverized fuel; IEA Clean Coal Center (2010) report CCC/168.
- [26] De Ris, J.: Fire Radiation A Review; 17th Symposium (International) on Combustion (1978) 1003-1016.
- [27] Denison, M. K.; Webb, B. W.: Development and Application of an Absorptionline Blackbody Distribution Function for CO<sub>2</sub>; International Journal of Heat and Mass Transfer 38 (1995) 1813-1821.
- [28] Denison, M. K.; Webb, B. W.: The Spectral Line–Based Weighted–Sum–of– Gray–Gases Model for H<sub>2</sub>O/CO<sub>2</sub> Mixtures; ASME Journal of Heat Transfer 117 (1995) 788-792.
- [29] Denison, M. K.; Webb, B. W.: The Spectral Line–Based Weighted–Sum–of– Gray–Gases Model in Nonisothermal Nonhomogenous Media; ASME Journal of Heat Transfer 117 (1995) 359-365.
- [30] Edwards, D. K., Balakrishnan, A.: Thermal Radiation by Combustion Gases; International Journal of Heat Transfer 16 (1973) 25-40.
- [31] Edwards, D. K., Menard, W. A.: Comparison of Models for Correlation of Total Band Absorption; Applied Optics 3 (1964) 621-625.

- [32] Erfurth, J.; Toporov, D.; Förster, M.; Kneer, R.: Simulation der Strahlungswärmeübertragung in einem Oxycoal–Großdampferzeuger; VDI-Berichte 2056, 24. Deutscher Flammentag, VDI Verlag Düsseldorf (2009) 139-144.
- [33] Farag, I. H., Allan, T. A.: Gray–Gas Approximation of Carbon Dioxide Standard Emissivity; ASME Journal of Heat Transfer 103 (1981) 403-405.
- [34] Farag, I. H.: Non–Luminous Gas Radiation Approximate Emissivity Models; Proceedings of the 7th International Heat Transfer Conference 2 (1982) 487-492.
- [35] Felske, J. D.; Tien, C. L.: The Use of the Milne–Eddington Absorption Coefficient for Radiative Heat Transfer in Combustion Systems; ASME Journal of Heat Transfer 99 (1977) 458-465.
- [36] Field, M. A.: Rate of Combustion of Size–Graded Fractions of Char from a Low Rank Coal between 1200 K – 2000 K; Combustion and Flame 13 (1969) 237-252.
- [37] Fiveland, W. A.: A Discrete Ordinates Method For Predicting Radiative Heat Transfer in Axisymmetric Enclosures; ASME paper no. 82–HT–20 (1982).
- [38] Fiveland, W. A.: Discrete–Ordinates Solutions of the Radiative Transport Equation for Rectangular Enclosures; ASME Journal of Heat Transfer 106 (1984) 699-706.
- [39] Fiveland, W. A.: Three–Dimensional Heat–Transfer Solutions by the Discrete–Ordinates Method; AIAA Journal of Thermophysics and Heat Transfer 2 (1988) 309-316.
- [40] Fleck, J. A.: The Calculation of Nonlinear Radiation Transport by a Monte Carlo Method: Statistical Physics; Methods in Computational Physics 1 (1961) 43-65.
- [41] ANSYS FLUENT 12.0, User's Guide; ANSYS Inc. (2009).
- [42] Foster, P. J.; Howarth, C. R.: Optical Constants of Carbons and Coals in the Infrared; Carbon 6 (1968) 719-729.

- [43] Fritsche, H.: Untersuchungen zur pneumatischen Förderung von Kohlenstaub zum Einblasen in den Hochofen unter besonderer Berücksichtigung von instabilen Förderzuständen; Dissertation, Technische Universität Clausthal (2013).
- [44] Grosshandler, W. L.: RADCAL: A Narrow–Band Model for Radiation Calculations in a Combustion Environment; NIST Technical Note 1402 (1993).
- [45] Hadvig, S.: Gas Emissivity and Absorptivity: A Thermodynamic Study; Journal of the Institute of Fuel 43 (1970) 129-135.
- [46] Hartmann, J. M., Levi Di Leon, R., Taine, J.: Line-by-Line and Narrow Band Statistical Model Calculations for H<sub>2</sub>O; Journal of Quantitative Spectroscopy and Radiative Transfer 32 (1984) 119-127.
- [47] Helble, J. J.; Sarofim, A. F.: Aggregate Formation from Vaporized Ash During Pulverized Coal Combustion; 21st Symposium (International) on Combustion (1986) 411-417.
- [48] Hottel, H. C.; Cohen, E. S.: Radiant Heat Exchange in a Gas–Filled Enclosure: Allowance for Nonuniformity of Gas Temperature; A.I.Ch.E. Journal 4 (1958) 3-14.
- [49] Hottel, H. C.; Sarofim, A. F.: Radiative Transfer; McGraw-Hill, New York, USA (1967).
- [50] Howell, J. R.: Calculation of Radiant Heat Exchange by the Monte Carlo Method; Technical Preprint prepared for Winter Annual Meeting of the American Society of Mechanical Engineers, Chicago, Illinois, USA (1965).
- [51] Howell, J. R.: The Monte Carlo Method in Radiative Heat Transfer; ASME Journal of Heat Transfer 120 (1998) 547-559.
- [52] Howell, J. R.; Perlmutter, M.: Monte Carlo Solution of Radiant Heat Transfer in a Nongrey Nonisothermal Gas with Temperature Dependent Properties; A.I.Ch.E. Journal (1964) 562-568.
- [53] Howell, J. R.; Perlmutter, M.: Monte Carlo Solution of Thermal Transfer Through Radiant Media Between Gray Walls; ASME Journal of Heat Transfer 86 (1964) 116-122.

- [54] IPCC: Special Report on Carbon Dioxide Capture and Storage; Prepared by Working Group III of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom, and New York, USA (2005).
- [55] IPCC: Summary for Policymakers; In: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom, and New York, USA (2007).
- [56] Johansson, R.; Leckner, B., Andersson, K.: Account For Ratios of H<sub>2</sub>O to CO<sub>2</sub> in the Calculation of Thermal Radiation of Gases with the Weighted– Sum–of–Grey–Gases Model; 6th Mediterranean Combustion Symposium (2009).
- [57] Johansson, R.; Leckner, B.; Andersson, K.; Johnsson, F.: Account For Variations in the H<sub>2</sub>O to CO<sub>2</sub> Molar Ratio When Modelling Gaseous Radiative Heat Transfer with the Weighted–Sum–Of–Grey–Gases Model; Combustion and Flame 158 (2011) 893-901.
- [58] Johnson, T. R.: Application of the Zone Method of Analysis to the Calculation of Heat Transfer from Luminous Flames; Ph. D. Thesis, University of Sheffield (1971).
- [59] Johnson, T. R.; Beer, J. M.: Radiative Heat Transfer in Furnaces: Further Development of the Zone Method of Analysis; 14th Symposium (International) on Combustion (1972) 639-649.
- [60] Kangwanpongpan, T.: Contribution to CFD Modeling of Lignite Oxy-fuel Combustion with Special Focus on Radiative Properties; Brandenburgische Technische Universität Cottbus, Dissertation (2013).
- [61] Kangwanpongpan, T.; França, F. H. R.; Corrêa da Silva, R.; Smith Schneider, P.; Krautz, H. J.: New correlations for the weighted–sum–of–gray–gases model in oxy–fuel conditions based on HITEMP 2010 database; International Journal of Heat and Mass Transfer 55 (2012) 7419-7433.
- [62] Kaß, H.; Tappe, S., Krautz, H. J.: The Combustion of Dry Lignite Under Oxy–Fuel Process Conditions in a 0.5 MWth Test Plant; Proceedings of the 9th International Conference on Greenhouse Gas Control Technologies (2009).

- [63] Kather, A.; Scheffknecht, G.: The Oxycoal Process with Cryogenic Oxygen Supply; Naturwissenschaften 96 (2009) 993-1010.
- [64] Khare, S. P.: Heat transfer in air-fired pulverized fuel furnaces retrofitted to oxy-fuel coal; University of Newcastle, Ph.D. Thesis (2008).
- [65] Khare, S.; Wall, T.: Radiative Transfer in Oxy–Fired Furnaces and Impact of Coal Properties; Technical note no. 27, Australian Black Coal Utilisation Research Ltd. (2007).
- [66] Kim, C.; Lior, N.: Easily computable good approximations for spectral radiative properties of particle–gas components and mixture in pulverized coal combustors; Fuel 74 (1995) 1891-1902.
- [67] Kim, T. K.; Smith, T. F.: Radiative and Conductive Transfer for a Real Gas in a Cylindrical Enclosure with Gray Walls; International Journal of Heat and Mass Transfer 28 (1985) 2269-2277.
- [68] Kluger, F.; Lysk, S.; Altmann, H.; Krohmer, B.; Stamatelopoulos, G.-N.: 30 MWth Oxyfuel Pilotanlage – Untersuchungsschwerpunkte und Auslegung des Dampferzeugers, 38. Kraftwerkstechnisches Kolloquium (2006).
- [69] Kobayashi, H.: Devolatilization of Pulverized Coal at High Temperatures; Massachusetts Institute of Technology, Ph.D. Thesis (1976).
- [70] Kobayashi, H.; Howard, J. B.; Sarofim, A. F.: Coal Devolatilization at High Temperatures; 16th Symposium (International) on Combustion (1977) 411-425.
- [71] Krishnamoorthy, G.; Sami, M.; Orsino, S.; Perera, A.; Shahnam, M.; Huckaby, E. D.: Radiation Modeling in Oxy–Fuel Combustion Scenarios; International Journal of Computational Fluid Dynamics 24 (2010) 69-82.
- [72] Krishnamoorthy, G.; Sami, M.; Orsino, S.; Perera, A.; Shahnam, M.; Huckaby, E. D.: Radiation Modeling in Oxy–Fuel Combustion Scenarios; Proceedings of the ASME Power Conference (2009).
- [73] Kuester, J. L.; Mize, J. H.: Optimization Techniques with FORTRAN; McGraw-Hill, New York, USA (1973).

- [74] Kunugi, M.; Jinno, H.: Determination of Size and Concentration of Soot Particles in Diffusion Flames by a Light–Scattering Technique; 11th Symposium (International) on Combustion (1967) 256-266.
- [75] Lallemant, N.; Sayre, A.; Weber, R.: Evaluation of Emissivity Correlations For H<sub>2</sub>O-CO<sub>2</sub>-N<sub>2</sub>/Air Mixtures and Coupling with Solution Methods of the Radiative Transfer Equation; Prog. Energy Combust. Sci. 22 (1996) 543-574.
- [76] Lallemant, N.; Weber, R.: Radiative Property Models for Computing Non-Sooty Natural Gas Flames, Part 1: Report on Radiation Modelling, IFRF Doc. no. G 08/Y/2 (1993).
- [77] Lallemant, N.; Weber, R.: Radiative Property Models For Computing Non-Sooty Natural Gas Flames, Part 2: Appendices, IFRF Doc. no. G 08/Y/2 (1993).
- [78] Laurendeau, N. M.: Heterogeneous Kinetics of Coal Char Gasification and Combustion; Progress in Energy and Combustion Science 4 (1978) 221-270.
- [79] Leckner, B.: Spectral and Total Emissivity of Water Vapor and Carbon Dioxide; Combustion and Flame 19 (1972) 33-48.
- [80] Lee, P. Y. C.; Hollands, K. G. T.; Raithby, G. D.: Reordering the Absorption Coefficient within the Wide Band for Predicting Gaseous Radiant Exchange; ASME Journal of Heat Transfer 118 (1996) 394-400.
- [81] Lee, S. C.; Tien, C. L.: Optical constants of soot in hydrocarbon flames; 18th Symposium (International) on Combustion (1981) 1159-1166.
- [82] Leiser, S.: Numerical Simulation of Oxy-fuel combustion; Universität Stuttgart, Dissertation (2010).
- [83] Liu, F.: Numerical Solutions of Three–Dimensional Non–Grey Gas Radiative Transfer Using the Statistical Narrow–Band Model; Journal of Heat and Mass Transfer 121 (1999) 200-203.
- [84] Liu, H.; Qiu, J.; Wang, X.; Sheng, C.; Kong, F.; Wu, H.: Radiative Heat Transfer Calculation of Coal–Fired Boiler in Oxy–Fuel Combustion; American Chemical Society Division of Fuel Chemistry Preprints 53 (2008) 510-511.
- [85] Liu, Y.; Rogg, B.: Prediction of Radiative Heat Transfer in Laminar Flames; Combustion Science and Technology 118 (1196) 127-145.

- [86] Lockwood, F. C.; Shah, N. G.: A New Radiation Solution Method for Incorporation in General Combustion Prediction Procedures; 18th Symposium (International) on Combustion (1981) 1405-1414.
- [87] Lockwood, F. C.; Rizvi, S. M. A.; Shah, N. G.: Comparitive Predictive Experience of Coal Firing; Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science (1986) 79-87.
- [88] Ludwig, C. B., Malkmus, B., Reardon, J. E., Thompson, J. A.: Handbook of Infrared Radiation from Combustion Gases; NASA Technical Report SP3080 (1973).
- [89] Marin, O.; Buckius, R. O.; Raithby, G. D.: Wide–Band Correlated–k Method Applied to Absorbing, Emitting and Scattering Media; Journal of Thermophysics and Heat Transfer 10 (1996) 364-371.
- [90] Marzouk, O. A.; Huckaby, E. D.: Nongray EWB and WSGG Radiation Modeling in Oxy–Fuel Environments; in: Zhu, J. (ed.): Computational Simulations and Applications, InTech, Rijeka, Croatia (2011).
- [91] Modak, A. T.: Radiation from Products of Combustion; Fire Safety Journal 1 (1979) 339-361.
- [92] Modest, M. F.: Radiative Heat Transfer; McGraw-Hill, New York, USA (1995).
- [93] Nakod, P.; Krishnamoorty, G.; Sami, M.; Orsino, S.: VA comparative evaluation of gray and non-gray radiation modeling strategies in oxy-coal combustion simulations; Applied Thermal Engineering 54 (2013) 422-432.
- [94] Nelder, J. A.; Mead, R.: A Simplex Method for Function Minimization; Computer Journal 7 (1965) 308-313.
- [95] Patch, R. W.: Effective Absorption Coefficients for Radiant Energy Transport in Nongrey, Nonscattering Gases; Journal of Quantitative Spectroscopy and Radiative Transfer 7 (1967) 611-637.
- [96] Phillips, W. J.: Band–Model Parameters for 4.3 μm CO<sub>2</sub> Band in the 300-1000 K Temperature Region; Journal of Quantitative Spectroscopy and Radiative Transfer 48 (1992) 91-104.

- [97] Phillips, W. J.: Band–Model Parameters of the 2.7 μm Band of H<sub>2</sub>O; Journal of Quantitative Spectroscopy and Radiative Transfer 43 (1990) 13-31.
- [98] Pierrot, L., Soufiani, A., Taine, J.: Accuracy of Narrow-Band and Global Models for Radiative Transfer in H<sub>2</sub>O, CO<sub>2</sub>, and H<sub>2</sub>/CO<sub>2</sub> Mixtures at High Temperature; Journal of Quantitative Spectroscopyand Radiative Transfer 62 (1999) 523-548.
- [99] Pierrot, L.; Rivière, P.; Soufiani, A.; Taine, J.: A Fictitious–Gas–Based Absorption Distribution Function Global Model for Radiative Transfer in Hot Gases; Journal of Quantitative Spectroscopy and Radiative Transfer 62 (1999) 609-624.
- [100] Queiroz, M.; Webb, B. W.: Particulate Behavior in a Controlled–Profile Coal–Fired Reactor: A Study of Coupled Turbulent Particle Dispersion and Thermal Radiation Transport, DOE–PC–91308 Final Report; Brigham Young University, Provo, UT 84602 (1996).
- [101] Rehfeldt, S.; Kuhr, C.; Schiffer, F.-P.; Weckes, P.; Bergins, C.: First Test Results of Oxyfuel Combustion with Hitachi's DST–burner at Vattenfall's 30 MWth Pilot Plant at Schwarze Pumpe, Energy Procedia 4 (2011) 1002-1009.
- [102] Rivière, P., Langlois, S., Soufiani, A., Taine, J.: An Approximate Database of H<sub>2</sub>O Infrared Lines For High Temperature Applications at Low Resolution. Statistical Narrow Band Model Parameters; Journal of Quantitative Spectroscopy and Radiative Transfer 53 (1995) 221-234.
- [103] Rivière, P., Soufiani, A., Taine, J.: Correlated–k and Fictitious Gas Methods for H<sub>2</sub>O near 2.7 μm; Journal of Quantitative Spectroscopy and Radiative Transfer 48 (1992) 187-203.
- [104] Rosseland, S.: Astrophysik auf atomtheoretischer Grundlage; Berlin (1931).
- [105] Rothman, L., Gamache, R. R., Tipping, R. H., Rinsland, R. H., Smith, C. P., Chris Benner, M. A. H., Malathi Devi, D., Flaud, D., Camy-Peyret, J. M., Perrin, C., Goldman, A., Massie, S. T. and Brown, L. R.: The HITRAN Molecular Database: Editions of 1991 and 1992; Journal of Quantitative Spectroscopy and Radiative Transfer 48 (1992) 469-507.
- [106] Rothman, L.; Gordon, I.; Barber, R.; Dothe, H.; Gamache, R.; Goldman, A.; Perevalov, V.; Tashkun, S.; Tennyson, J.: HITEMP, The High–Temperature

Molecular Spectroscopic Database; Journal of Quantitative Spectroscopy and Radiative Transfer 48 (2010) 2139-2150.

- [107] Sahajwalla, V.; Eghlimi, A.; Farrell, K.: Numerical Simulation of Pulverised Coal Combustion; International Conference on CFD in Mineral and Metal Processing and Power Generation (1997) 197-204.
- [108] Santos, S.; Haines, M.; Davison, J.: Challenges in the Development of Oxy– Combustion Technology For Coal–Fired Power Plant; Proceedings of the 31st International Technical Conference on Coal Utilization & Fuel Systems 1 (2006) 1-11.
- [109] Sarofim, A. F.; Howard, J. B.; Padia, A. S.: The Physical Transformation of the Mineral Matter in Pulverized Coal under Simulated Combustion Conditions; Combustion Science and Technology 16 (1977) 187-204.
- [110] Schack, K.: Berechnung der Strahlung von Wasserdampf und Kohlendioxid; Chemie Ingenieur Technik 42 (1970) 53-104.
- [111] Scheffknecht, G.; Al-Makhadmeh, L.; Schnell, U.; Maier, J.: Oxy-fuel coal combustion — A review of the current state-of-the-art; International Journal of Greenhouse Gas Control (2011), doi:10.1016/j.ijggc.2011.05.020.
- [112] Scutaru, D., Rosenmann, L., Taine, J.: Approximate Intensities of CO<sub>2</sub> hot bands at 2.7, 4.3 and 12 μm for High Temperature and Medium Resolution Applications; Journal of Quantitative Spectroscopy and Radiative Transfer 52 (1994) 765-781.
- [113] Selçuk, N.: Exact Solutions of Radiative Heat Transfer in Box–Shaped Furnaces; ASME Journal of Heat Transfer 107 (1985) 663-669.
- [114] Selçuk, N.; Siddall, R. G., Beér, J. M.: A Comparison of Mathematical Models of the Radiative Behaviour of a Large–Scale Experimental Furnace; 16th Symposium (International) on Combustion (1977) 53-62.
- [115] Selçuk, N.; Tahiroğlu, Z.: Exact Numerical Solutions for Radiative Transfer in Cylindrical Furnaces; International Journal of Numerical Methods in Engineering 26 (1988) 1201-1212.
- [116] Shah, N. G.: New Method of Computation of Radiation Heat Transfer in Combustion Chambers; Ph. D. Thesis, Imperial College of Science and Technology London (1979).

- [117] Shih, T.-H.; Lion, W. W.; Shabbir, A.; Yang, Z.; Zhu, J.: A New k-ε Eddy– Viscosity Model for High Reynolds Number Turbulent Flows — Model Development and Validation; Computer Fluids 24, Issue 3 (1995) 227-238.
- [118] Siegel, R.; Howell, J. R.: Thermal Radiation Heat Transfer, 4th Edition; Francis & Taylor, New York, USA (2002).
- [119] Sims, R. E. H.; Schock, R. N.; Adegbululgbe, A.; Fenhann, J.; Konstantinaviciute, I.; Moomaw, W.; Nimir, H. B.; Schlamadinger, B.; Torres-Martínez, J.; Turner, C.; Uchiyama, Y.; Vuori, S. J. V.; Wamukonya, N.; Zhang, X.: Energy Supply; In: Climate Change 2007: Mitigation. Contribution of Working Group III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom, and New York, USA (2007).
- [120] Smith, I. W.: The Intrinsic Reactivity of Carbons to Oxygen; Fuel 57 (1978), 409-414.
- [121] Smith, I. W.: The Combustion Rates of Coal Chars; 19th Symposium (International) on Combustion (1982) 1045-1065.
- [122] Smith, T. F.; Shen, Z. F.; Friedman, J. N.: Evaluation of Coefficients for the Weighted Sum of Gray Gases Model; ASME Journal of Heat Transfer 104 (1982) 602-608.
- [123] Smyth, K. C.; Shaddix, C. R.: The elusive history of m = 1.57 0.56i for the refractive index of soot; Combustion and Flame 107 (1996) 314-320.
- [124] Snegirev, A. Y.: Monte Carlo Modeling of Radiative Heat Transfer in Unconfined Buoyant Turbulent Diffusion Flames; Proceedings of Eurotherm Seminar 73 (2003) 259-270.
- [125] Soufiani, A., Hartmann, J. M., Taine, J.: Validity of Band Model Calculations for CO<sub>2</sub> and H<sub>2</sub>O Applied to Radiative Properties and Conductive– Radiative Transfer; Journal of Quantitative Spectroscopy and Radiative Transfer 33 (1985) 243-257.
- [126] Steward, F. R.; Kocaefe, Y. S.: Total Emissivity and Absorptivity Models for Carbon Dioxide, Water Vapor and their Mixtures; Proceedings of the 8th Heat Transfer Conference (1986) 735-740.

- [127] Stieß, M.: Mechanische Verfahrenstechnik 2; Springer, Berlin, Heidelberg, New York (1994).
- [128] Ströhle, J.: Spectral Modelling of Radiative Heat Transfer in Industrial Furnaces; Universität Stuttgart, Dissertation (2003).
- [129] Strömberg, L.; Lindgren, G.; Anheden, M.; Simonsson, N.; Köpcke, M.: Vattenfall's 30 MWth Oxyfuel Pilot Plant, Proceedings of the 8th International Conference on Greenhouse Gas Control Technologies (2006).
- [130] Strömberg, L.; Lindgren, G.; Jacoby, J.; Giering, R.; Anheden, M.; Burchhardt, U.; Altmann, H.; Kluger, F.; Stamatelopoulos, G.-N.: Update on Vattenfall's 30 MWth Oxyfuel Pilot Plant in Schwarze Pumpe, Energy Procedia 1 (2009) 581-589.
- [131] Taine, J., Soufiani, A.: From Spectroscopic Data to Approximate Models; Advances in Heat Transfer 33 (1999) 295-414.
- [132] Tappe, S., Krautz, H. J.: Experimental Investigations of Combustion Behaviour in Various O<sub>2</sub>/CO<sub>2</sub>-Atmospheres; 4th International Conference on Clean Coal Technologies and 3rd International Freiberg Conference on IGCC and XtL Technologies (2009).
- [133] Taylor, P. B., Foster, P. J.: Some Gray Gas Weighting Coefficients for CO<sub>2</sub>– H<sub>2</sub>O Soot Mixtures; International Journal of Heat and Mass Transfer 18 (1975) 1331-1332.
- [134] Taylor, P. B., Foster, P. J.: The Total Emissivities of Luminous and Non– Luminous Flames; International Journal of Heat and Mass Transfer 17 (1974) 1591-1605.
- [135] Tien, C. L.; Lee, S. C: Flame Radiation; Progress in Energy and Combustion Science 8 (1982) 41-59.
- [136] Tigges, K.-D.; Klauke, F.; Bergins, C.; Busekrus, K.; Niesbach, J.; Ehmann, M.; Kuhr, C.; Hoffmeister, F.; Vollmer, B.; Buddenberg, T.; Wu, S.; Kukoski, A.: Conversion of Existing Coal–Fired Power Plants to Oxyfuel Combustion: Case Study with Experimental Results and CFD–Simulations; Energy Procedia 1 (2009) 549-556.

- [137] Traugott, S. C.: On Grey Absorption Coefficients in Radiative Transfer; Journal of Quantitative Spectroscopy and Radiative Transfer 8 (1968) 971-999.
- [138] Upham, P.; Roberts, T.: Public Perceptions of CCS in Context: Results of NearCO<sub>2</sub> Focus Groups in the UK, Belgium, the Netherlands, Germany, Spain and Poland; Energy Procedia 4 (2011) 6338-6344.
- [139] Viskanta, R.; Mengüç, M. P.: Radiation Heat Transfer in Combustion Systems; Progress in Energy and Combustion Science 13 (1987) 97-160.
- [140] Viskanta, R.; Ugnan, A.; Mengüç, M. P.: Predictions of Radiative Properties of Pulverized Coal and Fly–Ash Polydispersions; ASME paper no. 81–HT–24 (1981).
- [141] Wall, T.; Liu, Y.; Spero, C.; Elliot, L.; Khare, S.; Rathnam, R.; Zeenathal, F.; Moghtaderi, B.; Buhre, B.; Sheng, C.; Gupta, R.; Yamada, T.; Makino, K.; Yu, J.: An Overview on Oxyfuel Coal Combustion — State of the Art Research and Technology Development; Chemical Engineering Research and Design 87 (2009) 1003-1016.
- [142] Woycenko, D. M.; Ikeda, I.; Van De Kamp, W. L.: Combustion of Coal in A Mixture of Oxygen and Recycled Flue Gas; IFRF Doc. no. F98/Y/1 (1994).
- [143] Yin, C.; Johansen; L. C. R.; Rosendahl, L. A.; Kær, S. K.: New Weighted Sum of Gray Gases Model Applicable to Computational Fluid Dynamics (CFD) Modeling of Oxy–Fuel Combustion: Derivation, Validation, and Implementation; Energy Fuels 24 (2010) 6275-6282.
- [144] Zhang, H.; Modest, M. F.: Full–Spectrum k–Distribution Correlations for Carbon Dioxide Mixtures; Journal of Thermophysics and Heat Transfer 17, No. 2 (2003) 259-263.