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Research Paper

Radiative effects of carbon monoxide in high-temperature carbon dioxide plumes and implications for modeling thermal plasmas

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ABSTRACT

High-temperature heating processes that are transitioning towards low-carbon operations are emphasizing work on gas radiation modeling. The formation of CO from CO₂-dissociation is significant at high temperatures, raising questions about the relative importance of CO in broadband gas radiation models. This work focuses on the radiative effect of CO in high-temperature CO₂-plumes. The analysis combines speciation in these zones with the construction of a framework to evaluate the radiative contribution of gases. It is shown that under peak temperature conditions associated with thermal CO₂-plasma plumes, where CO₂-dissociation and the formation of CO are significant, the chemical timescales are sufficient to approximate equilibrium. Within this thermochemical and optical regime, CO₂ dominates the radiative contribution, as compared to CO. The reason for this is the extensive spectral overlap in the relevant infrared bands between the two species. For temperatures of approximately 1200–3000 K and CO/CO₂ ratios of 1 or below, treating CO as transparent results in less than 6% error in the peak radiative source term and about 7% error in gas-mixture emissivity. The radiative contribution of CO becomes more pronounced under CO-rich conditions, and at temperatures above approximately 3800 K, where CO₂ dissociation increases the relative importance of CO. Thus, the results provide a quantitative basis for effectively modeling CO as a transparent gas in broadband representations of CO-CO₂ mixtures, thereby simplifying the treatment of CO in radiation models within the identified thermochemical and optical regime relevant for novel applications such as CO₂-based thermal plasmas.

1. Introduction

Thermal radiation, which is the dominant heat transfer mechanism in high-temperature processes, is influenced by the participating gases and particles. In conventional combustion systems, particles such as unburnt fuel, ash, and soot dominate the radiative heat transfer, particularly in high-temperature applications, such as rotary kilns, (cylindrical rotating furnaces used for material processing) [1,2]. These processes are currently being investigated for low-carbon alternatives, whereby they can transition from fossil fuel-based heating to electrification. Thermal plasma systems offer such an alternative, providing a carbon-free heat source that is characterized by higher peak temperatures and increased gas concentrations, depending on the selected working gas. Under these conditions, gas radiation becomes the primary contributor to radiative heat transfer, emitting in specific wavelength bands that reflect the temperature, species partial pressures, and optical path length, such that complex models are needed for accurate predictions. As shown in a previous work [3], the conditions relevant to

thermal plasma systems necessitate careful selection of radiation models, since gas radiation becomes a significant determinant of the thermal field.

The weighted-sum-of-gray-gases model (WSGGM) is a commonly used broadband gas radiation model that is favored due to its high predictive accuracy relative to its computational efficiency. Consequently, the WSGGM is often employed in commercial computational fluid dynamics (CFD) simulations, as in our previous studies [3–5]. The WSGGM is an absorption coefficient model that considers the entire gas spectrum and aims to fit the radiative properties of a gas mixture within a specified range of temperatures, gas mixtures, and pressure path lengths [6]. As a result, various parameters for WSGGM that are suitable for specific operating conditions have been reported [5,7–21]. The primary radiating species in these models, such as H₂O and CO₂, are typically found in combustion processes.

Our recent work [5] extended the applicable range of the WSGGM to temperatures up to 5000 K, including H₂O/CO₂ ratios in the range of 0.4–4.0 (relevant to conventional combustion conditions), as well as only H₂O/CO₂ in a transparent gas mixture at concentrations in the

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Nomenclature			
I	intensity	τ	transmissivity
S	pathlength	γ	mean line half-width
P	total pressure	ω	quadrature weight in a discrete direction
Y	gaseous molar fraction of species	μ	ordinate
T	temperature	ξ	normalized deviation by treating CO as transparent in source term predictions
k_k	mean line-intensity to typical line-spacing within a narrow band	ς	deviation by treating CO as transparent in gas-mixture emissivity predictions
d_k	inverse of mean line-spacing	<i>Subscript</i>	
∇q	radiative source term	ν	wavenumber/spectral property
N	total number of discrete directions	b	black body
s	the coordinates in a radiative path	n	cell number
S_m	distance between plates	m	discrete direction
<i>Greek symbols</i>		<i>Superscript</i>	
κ	absorption coefficient	+	absorbed
ϵ	emissivity	-	emitted

range of 0.05–1.0 (relevant to hydrogen combustion and thermal plasmas using H_2O/CO_2 as a working gas).

Under high-temperature conditions, the level of formation of CO from CO_2 -dissociation is significant, raising questions on the relevance of including the radiative contribution of CO to the total radiation in gas radiation modeling. While several studies have focused on developing WSGGM parameters that incorporate CO into their models, these conditions are often limited to oxyfuel combustion processes, where the concentrations of CO are expected to be significant, especially in proximity to the burner region of the flame [22], or under gasification conditions [9,23]. For example, Yin et al. [12,13] and Wu et al. [12,13] have developed modified WSGGM parameters that incorporate CO, demonstrating using CFD simulations of oxy-natural gas-fired furnaces the impacts of high levels of CO on radiation and on predicting the temperature distribution, as compared with conventional radiation models. The modified WSGGM [12] applies to CO mole fractions ranging from 0 to 0.45, H_2O/CO_2 ratios ranging from 0.01 to 4, and temperatures up to 3000 K.

Although these studies provide radiation models that account for CO, they primarily address the accurate representation of CO as a participating species rather than the conditions under which its radiative contribution can be neglected. The role of CO as a radiating species under high-temperature CO_2 -dissociation conditions, such as those related to thermal plasmas using CO_2 as the working gas, remains limited. In these systems, the thermal field can be divided into two zones: a high-temperature zone that is affected by CO_2 -dissociation; and a lower-temperature zone that is influenced by CO oxidation. The strong coupling between species composition and spectral radiation under these conditions introduces additional complexities in relation to assessing the relative significance of CO in a CO_2 -mixture in gas radiation modeling. Some recent studies provide relevant context for CO_2 -rich thermal plasma environments, including hybrid plasma systems with H_2O and complex feedstocks [24,25]. However, these systems differ in the working gas compositions and process conditions from those considered here. This study instead focuses on isolating the CO effects of radiation in CO_2 -CO-air mixtures relevant to the high-temperature CO_2 -plasma environments studied in [26,27].

This work evaluates and analyzes the radiative effects of CO under conditions associated with CO_2 -mixtures affected by high-temperature chemistry. Specifically, this work identifies the thermochemical and optical conditions under which CO can be treated as a transparent gas with limited impact on predicted radiation. A modeling approach that involves a combination of detailed chemistry, radiation, and computational fluid dynamics—incorporating descriptions of chemistry, radiative

transfer, and turbulence, respectively—is employed in the analysis. As a representative application, the conditions for a CO_2 -plume under thermal plasma heating are considered.

2. Methodology

The analysis focuses on the radiating species CO_2 and CO under conditions that are representative of high-temperature CO_2 -plumes. It includes three model frameworks (Fig. 1): (i) a chemistry framework to isolate and describe in detail the high-temperature chemistry in CO_2 -mixtures; (ii) a radiation framework to assess the local radiative contributions of CO_2 and CO, as well as the impact on predicted radiation for treating CO as transparent, through detailed radiative transfer calculations; and (iii) the application of these findings to a CO_2 -plasma-heated kiln utilizing a computational fluid dynamics (CFD) framework that incorporates chemistry, radiative transfer, and turbulence. This framework is derived from previous studies [26,27]. The inputs to the chemistry framework are user-defined and include temperature, pure CO_2 , CO_2 -air mixtures, and CO_2 -CO-air mixtures. The outputs consist of the corresponding species composition of the mixture based on the chemistry prediction and the residence time needed for the mixture to relax to chemical equilibrium. The inputs to the radiation framework include both user-defined profiles and profiles generated from the chemistry and CFD frameworks to predict the radiative intensity. The inputs to the CFD framework are the operating conditions [26,27], defined as boundary conditions, and the outputs are the predicted flow field including the distributions of gas composition and temperature, as well as the residence time of the flow within the hot CO_2 -plume. The specification of each framework is given in the following sections.

2.1. Chemistry framework

The chemistry framework predicts the high-temperature chemistry and the corresponding species concentrations at various temperatures, and compares these concentrations to the chemical equilibrium. As such, the framework consists of chemical equilibrium calculations and finite-rate plug flow reactor (PFR) simulations. The gas-phase kinetics and thermodynamic properties are described according to the reaction extracted from the GRI 3.0 mechanisms [28], as expressed in reaction R1, which represents the dissociation/oxidation of CO_2/CO . The formation of CO_2/CO from other reaction sources is considered outside the scope of this study. The analysis includes calculations made on CO_2 -mixtures under isothermal and non-isothermal conditions. Under isothermal conditions, the temperature dependence of the chemical

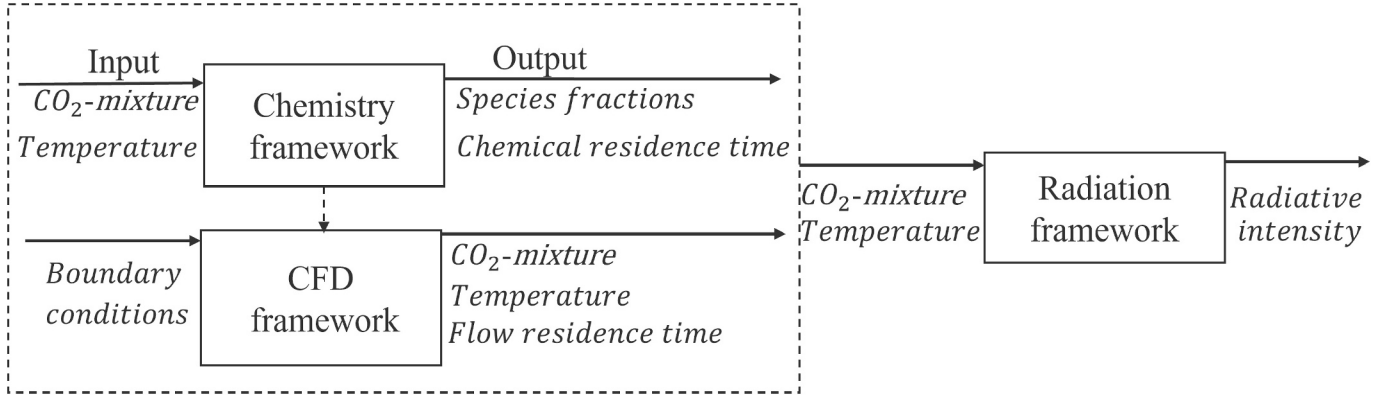


Fig. 1. The model frameworks used herein include detailed chemistry and radiation frameworks, along with a computational fluid dynamics framework that incorporates turbulence and lower levels of complexity in the descriptions of the chemistry and radiation.

kinetics is isolated by maintaining a fixed temperature across the system. Case 1 in Table 1 analyzes the radiative effects (predicted by the radiation framework) of pure CO_2 under isothermal temperatures (300–5000 K), where the species composition is affected by dissociation, as predicted by the chemistry framework under chemical equilibrium conditions. For the remaining cases (Cases 2 and 3), the selected temperature inputs are 2100 K, 2500 K, and 3000 K, since dissociation of pure CO_2 occurs above 2000 K under chemical equilibrium conditions, as will be shown in the Results section. For Case 2, the calculations aim to evaluate the chemical timescales required for the species to relax towards their corresponding chemical equilibrium values at the different temperatures. Specifically, the aim is to show the timescale needed to reach chemical equilibrium when a pure CO_2 stream is affected by dissociation. Under non-isothermal conditions, the coupling between chemistry and thermal evolution allows dissociation/oxidation to influence directly the system temperature. Case 3 evaluates the chemical timescale that a reacting mixture needs to relax towards chemical equilibrium. The different cases are summarized in Table 1.



To determine whether the chemical timescales predicted by the chemistry framework are sufficiently short to justify the assumption of chemical equilibrium under the studied conditions, they are compared to a characteristic flow timescale obtained from the CFD framework. The flow timescale is estimated by tracking a mass-less tracer particle injected at the plasma torch outlet, thereby calculating its residence time within the hot CO_2 -plume, corresponding to the primary reaction zone. This zone extends for approximately 40 cm, retaining temperatures between 3000 K and 2100 K. The residence time of the tracer particle

Table 1

Cases 1–3 employing the Chemistry framework for various input temperatures and CO_2 -mixtures. The output is either the radiative intensity predicted by the radiation framework or the chemical residence time.

Case	Input: Temperature [K]			Input: Composition [vol%]	Output
1: Chemistry framework: CO_2 -mixtures	300–5000			From Chemistry framework	Radiative intensity
2: Isothermal	2100	2500	3000	100% CO_2	Residence time to relax to chemical equilibrium
3: Non-isothermal	2100	3000		50% CO_2 & 50% air 25% CO_2 & 25% CO & 50% air	Residence time to relax to chemical equilibrium

along this trajectory is 2 ms; however, the time may vary for different plasma torch configurations. The particle residence times as a function of the distance from the plasma torch outlet, along with the corresponding temperature profiles, are provided in the Supplementary Material (Fig. S1).

2.2. Radiation framework

The radiation framework, which requires temperature and species concentrations as inputs, calculates the radiative intensity by the radiative transfer equation (RTE) that describes, for a set direction, s , the radiative intensity change in the spectral range, ν , from the summed contribution of the absorption, emission, and scattering mediated by the participating gases and particles. If only gases and/or soot are considered, the radiative intensity can be determined by discretizing the radiative transfer equation over control volumes, n , and in terms of transmissivities, in accordance with Eq. (1):

$$\bar{I}_{\nu,n} = \bar{I}_{\nu,n-1} \bar{\tau}_{\nu,n-1 \rightarrow n} + \bar{I}_{b,\nu,n-1/2} (1 - \bar{\tau}_{\nu,n-1 \rightarrow n}) \quad (1)$$

Here, the radiative intensity is modeled using a discrete transfer model (DTM) [1], and is suitable for evaluating the RTE in simple geometries, as it is computationally demanding for multidimensional geometries. In the DTM, the RTE is calculated along a set of rays, each paired with a corresponding quadrature weight, to span the angle range of 4π , and each ray is traced from its source of origin through the computational domain.

For the gas radiation calculations, the radiation framework applies a statistical narrow-band model (SNBM), which divides the gas spectrum into spectral wavebands consisting of a range of wavelengths. Eq. (2) describes the spectral-averaged transmissivity for a waveband, k , determined by the SNBM. Here, the SNBM applies the Malkmus model [29], with band parameters acquired from the work of Rivière and Soufiani [30], valid for temperatures up to 5000 K. The SNBM is selected in the radiation model because it performs as accurately as line-by-line models [31].

$$\bar{\tau}_{\nu_k} = \exp \left[-2\gamma / d_k \left((1 + YPSk d_k / \gamma)^{1/2} - 1 \right) \right] \quad (2)$$

For a mixture of gas species, the total transmissivity is the product of the individual gas species transmissivities, as expressed in Eq. (3), and the emissivity is a function of the average black body intensity of band k and the total transmissivity, as expressed in Eq. (4).

$$\bar{\tau}_{\nu_{k,\text{gas}}} = \prod_{\text{species}} \bar{\tau}_{\nu_{k,\text{specie}}} \quad (3)$$

$$\varepsilon_{\text{SNBM}} = 1 - 1 / I_b \sum_k \bar{I}_{b\nu_k} \bar{\tau}_{\nu_k} \quad (4)$$

The bands modeled by the SNBM [30] include the major infrared absorption bands of CO_2 , characterized by prominent bands centered around 667 cm^{-1} ($15 \mu\text{m}$), 2350 cm^{-1} ($4.3 \mu\text{m}$), and 3660 cm^{-1} ($2.7 \mu\text{m}$), together with weaker bands close to 960 cm^{-1} ($10.4 \mu\text{m}$), 1060 cm^{-1} ($9.4 \mu\text{m}$), and 5200 cm^{-1} ($2.0 \mu\text{m}$) [6]. For CO , the dominant infrared absorption band occurs near 2143 cm^{-1} ($4.66 \mu\text{m}$), with a weaker band at around 4260 cm^{-1} ($2.34 \mu\text{m}$). Fig. 2 illustrates an example of the influences of the major infrared bands with increasing temperature (1000 K , 2000 K , and 3000 K), using the SNBM. A pressure path length of 0.01 atm m is selected to avoid saturating the bands, thereby making the overall absorption magnitude of the bands relative to radiative intensity more comparable. The sensitivity of the selected pressure path length will be presented in the *Results* section. In Fig. 2a, the primary CO_2 emission bands appear close to 2350 cm^{-1} and 3660 cm^{-1} and are the main contributors to the total gas emissivity under these conditions. Although the 667 cm^{-1} band is spectroscopically strong [6], its contribution to the emitted intensity is comparatively small within the increased temperature range considered ($2000\text{--}3000\text{K}$). In Fig. 2b, the primary CO emission occurs near 2143 cm^{-1} and this is the main contributor to the radiative properties of CO at these temperatures, whereas a weaker band close to 4260 cm^{-1} contributes to a lesser extent. Overall, Fig. 2 shows that both CO_2 and CO exhibit dominant emission features within a similar spectral region between approximately 2100 cm^{-1} and 2300 cm^{-1} . The resulting spectral overlap between these bands suggests that CO_2 , which is observed to be a stronger radiative emitter than CO (note the scale difference on the y-axis), partially suppresses the radiative contribution of CO in CO_2 - CO mixtures under high-temperature conditions.

In CFD, radiative transfer is included as a source term in the energy transport equation, determined by locally absorbed and emitted radiative energy levels. In this work, the radiation framework is employed to predict the radiative source term in detail along a path within a 1D gas domain that consists of two infinite black plates. The framework allows for analyses of the radiative contributions of CO_2 and CO . The radiative source term is the divergence of the radiative flux, q , as determined from the radiative intensity, I , ordinate, μ , and quadrature weight, ω , in the discrete direction, m , as expressed in Eq. (5):

$$\nabla q = \sum_{m=1}^{N/2} \omega_m \mu_m (I_n^+ - I_n^-) \quad (5)$$

The analysis includes prescribed user-defined temperature and gas distributions by varying the temperature and CO_2/CO ratios according to cosine profiles. The selected profiles in Cases 4 and 5 in Table 2 represent rich CO_2/CO mixture conditions at temperature levels that are similar to those of the CO_2 -plume reported in the CFD framework [27]. The profiles in Cases 6–8 in Table 2 are inspired by the temperature levels and CO_2/CO ratios reported for propane–air combustion and oxy-

Table 2

Descriptions of Cases 4–10 that employ the radiation framework for various input temperatures and CO_2 - CO profiles corresponding to conditions similar to those reported previously [27] [32] [33]. The output is either the radiative source term along the 1D domain or the radiative intensity. The case numbers follow the previously defined chemistry framework cases.

Case	Input: Temperature [K]	Input: Composition [vol%]	Output
4. Rich CO_2 -mixture	2,000 – $1,000\cos(2\pi s/S_m)$	$Y_{\text{CO}_2} : 0.475 - 0.425\cos(2\pi s/S_m)$ $Y_{\text{CO}} : 0.075 - 0.025\cos(2\pi s/S_m)$	Source term
5. Rich CO -mixture	2,000 – $1,000\cos(2\pi s/S_m)$	$Y_{\text{CO}_2} : 0.075 - 0.025\cos(2\pi s/S_m)$ $Y_{\text{CO}} : 0.475 - 0.425\cos(2\pi s/S_m)$	Source term
6. Propane-air combustion [32]	1,425 – $425\cos(2\pi s/S_m)$	$Y_{\text{CO}_2} : 0.075 - 0.025\cos(2\pi s/S_m)$ $Y_{\text{CO}} : 0.075 - 0.025\cos(2\pi s/S_m)$	Source term
7. Propane-oxyfuel combustion [32]	1375 – $375\cos(2\pi s/S_m)$	$Y_{\text{CO}_2} : 0.9 - 0.05\cos(2\pi s/S_m)$ $Y_{\text{CO}} : 0.1 - 0.05\cos(2\pi s/S_m)$	Source term
8. Blast furnace conditions [33]	1,600 – $400\cos(2\pi s/S_m)$	$Y_{\text{CO}_2} : 0.055 - 0.045\cos(2\pi s/S_m)$ $Y_{\text{CO}} : 0.325 - 0.125\cos(2\pi s/S_m)$	Source term
9. CFD framework: CO_2 -plume [27]	From across a cross-section within the CFD framework	From across a cross-section within the CFD framework	Source term
10. CFD framework: CO_2 -plume [27]	From the centerline within the CFD framework	From the centerline within the CFD framework	Radiative intensity

fuel combustion with flue gas recirculation, in the work of Andersson and Johnsson [32], and blast furnace conditions, such as those reported by Fu et al. [33], although they are not intended to replicate these systems directly. The reason for using these cases is to analyze the influences of various temperature and CO_2/CO ratios on radiation. In all the user-defined cases, H_2O is neglected; although H_2O is considered an important participating species, this work focuses on analyzing exclusively the radiative effects of CO_2/CO in detail and the radiative impact of treating CO as transparent. The wall temperature is maintained at 1000 K , and the plate length, S_m , is 1 m . Furthermore, the analysis (Cases 9 and 10) includes a domain that is defined by a cross-section of a kiln diameter (0.58 m), incorporating the CFD-generated temperature and gas composition conditions reported previously [27]. Table 2

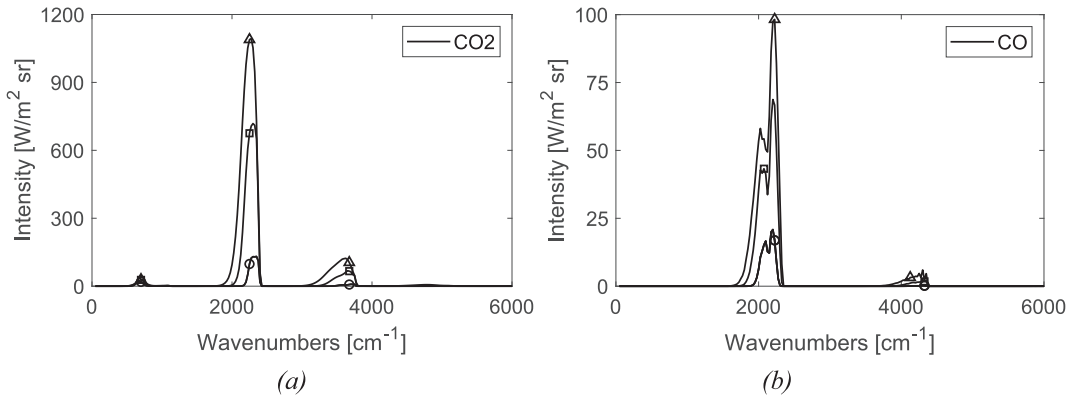


Fig. 2. Planck distributions of pure CO_2 (a) and CO (b) at 1000 K (circle), 2000 K (square), and $3,000 \text{ K}$ (triangle) using the SNBM. The pressure path length is 0.01 atm m .

summarizes the cases considered in the radiation framework.

The error introduced by treating CO as transparent for the radiative transfer calculations is determined as the difference between the calculated radiative source terms—or, in the case of spectral calculations, the difference in gas-mixture emissivity—according to Eq. (6). For the radiative source term error, the difference is normalized by the peak source term value to identify where the maximum error occurs.

$$\xi = |\nabla q_{CO_2+CO} - \nabla q_{CO_2}| / |\max(\nabla q_{CO_2+CO})|$$

$$\varsigma = (\epsilon_{CO_2+CO} - \epsilon_{CO_2}) / \epsilon_{CO_2+CO} \quad (6)$$

CFD framework.

The CFD framework is employed as an application to model a realistic system. The CFD model, described in [27] includes sub-models for combustion, turbulence, and radiation, to allow the resolution of sets of governing transport equations for mass, momentum, energy, and chemical species over a discretized computational domain, thereby generating profiles for temperature and species. These profiles are subsequently used as inputs to the radiation framework.

The CFD framework represents the pilot-scale kiln unit presented in previous studies [26,27]. The empty kiln is operated at 215 kW_{el} (159 kW_{th}) using an electrically powered thermal plasma, with CO_2 as the working gas, which enters at a 5° angle tilted towards the product feed. As the experimental conditions upon which the model is based suffered from air leakage, an air leakage boundary was included in the simulations, such that the simulated species comprise a CO_2 - CO -air mixture.

The framework has been verified against experimental measurements, including outlet O_2 and CO_2 concentrations obtained using FTIR and radiative heat flux measurements at available axial ports along the furnace length. The inner wall temperature measurements are acquired using a pyrometer, while the outer wall temperatures are measured with an infrared (IR) camera.

3. Results

The results presented in this section include the predictions of species concentration from the chemistry framework, the radiative contributions of CO_2 and CO and the radiative impact of treating CO as transparent from the radiation framework, and the simulated flow field obtained from the CFD framework in [27].

3.1. Predictions of CO_2 -mixtures affected by high-temperature chemistry

The chemistry framework is applied to calculate the species concentrations in a pure CO_2 -stream under isothermal chemical equilibrium conditions, i.e., in a situation where the CO_2 -stream is affected by dissociation, and chemical equilibrium is achieved at every temperature. Fig. 3 illustrates the radiative intensity (for Case 1) calculated using the radiation framework under these conditions for three scenarios:

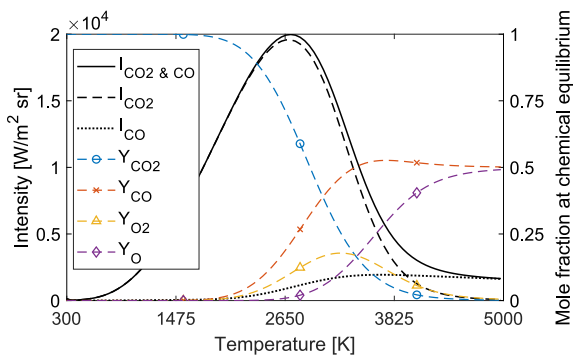


Fig. 3. Radiative intensities under various isothermal chemical equilibrium conditions (for Case 1).

combined CO_2 and CO radiation; CO_2 radiation only (with CO treated as transparent); and CO radiation only (with CO_2 treated as transparent). As the temperature increases, CO_2 progressively dissociates into CO , with the level of dissociation becoming significant at about 2100 K. The total radiative intensity peaks at a temperature close to 2700 K and thereafter decreases due to the reduction in CO_2 concentration, indicating that CO_2 is dominating in terms of radiative contribution. At sufficiently high temperatures, around 3850 K, CO is observed to become the dominant radiating species, although this occurs only when CO_2 is significantly depleted. The impact on radiation for a CO_2 -mixture under non-isothermal conditions is discussed below.

Table 3 summarizes the PFR simulation results for Cases 2 and 3 under various isothermal and non-isothermal conditions, to show the influence of temperature on chemistry. For Case 2 at 2100 K and for lower temperatures, the level of CO_2 -dissociation remains modest, and the system remains close to chemical equilibrium throughout the PFR. At higher temperatures (2500–3000 K), equilibrium is approximated at progressively shorter residence times, indicating a rapid relaxation towards equilibrium under these extreme temperature conditions. For Case 3, the temperature profile evolves from the initial temperature due to CO_2 -dissociation and CO -oxidation for mixtures of CO_2 and air, as well as for CO . For a CO_2 -air mixture at 2100 K, the dissociation of CO_2 again remains modest, and the system is close to chemical equilibrium throughout the PFR. At 3000 K, chemical equilibrium conditions are approximated at very short residence times. Including CO in the mixture requires longer residence times at lower temperatures (2100K), whereas at 3000 K the system relaxes towards equilibrium almost instantaneously. Overall, Cases 2 and 3 indicate that at the highest temperature levels, chemical equilibrium is approximated at residence times that are comparable to those estimated for the mass-less tracer particle through the high-temperature zone of the CFD-simulated CO_2 -plume (~2 ms). This suggests that the flow is likely to approach chemical equilibrium at the highest temperature level (3000K), but not at the lower levels (2100–2500 K). However, under these lower-temperature levels, the level of CO_2 -dissociation remains relatively low, and the CO concentrations are low. Consequently, the chemical equilibrium assumption is used to represent the species composition in the subsequent radiation analysis within the CO_2 -plume, rather than as an assertion that chemical equilibrium is fully attained in the plume. The radiation analysis of Cases 4–8 will demonstrate the effect of species compositions independently of the equilibrium assumption. The corresponding PFR simulation results for Cases 2 and 3 as a function of residence time are provided in the Supplementary Material (Figs. S2 and S3).

3.2. Radiative source term predictions for user-defined profiles

The radiative source term is predicted for Cases 4–8, employing user-defined temperature profiles and CO_2 / CO -mixtures, so as to be repre-

Table 3

Residence times needed for Cases 2 and 3 to relax to chemical equilibrium.

Case 2		
Temperature [K]	Composition [vol%]	Residence time needed to relax to chemical equilibrium [ms]
2100	100% CO_2	–
2500	100% CO_2	~10
3000	100% CO_2	~1.8
Case 3		
Temperature [K]	Composition [vol%]	Residence time needed to relax to chemical equilibrium [ms]
2100	50% CO_2 50% air	–
3000	50% CO_2 50% air	~2
2100	25% CO_2 25% CO 50% air	~10
3000	25% CO_2 25% CO 50% air	~1

sentative of rich CO_2/CO conditions at thermal plasma temperatures, and under propane-air and propane-oxyfuel combustion conditions, as well as under blast furnace conditions. As shown in Fig. 4a, at high CO_2/CO ratios (Case 4), CO exhibits a negligible impact on the total radiation. However, at significantly lower CO_2/CO ratios (Case 5), Fig. 4b shows a stronger influence of CO . In this case, treating CO as a transparent species, i.e., considering only CO_2 as the radiating species,

results in the lowest CO_2 contribution among the studied cases, corresponding to 88% of the combined CO_2 and CO radiative source term at the domain center, where both the temperature and CO concentration peak. Under propane-air combustion conditions (Case 6), comprised of comparable CO_2/CO ratios, Fig. 4c shows a modest impact of CO on the total radiation, with the maximum effect noted at the domain center. In Fig. 4d, representing propane-oxyfuel combustion (Case 7), the high

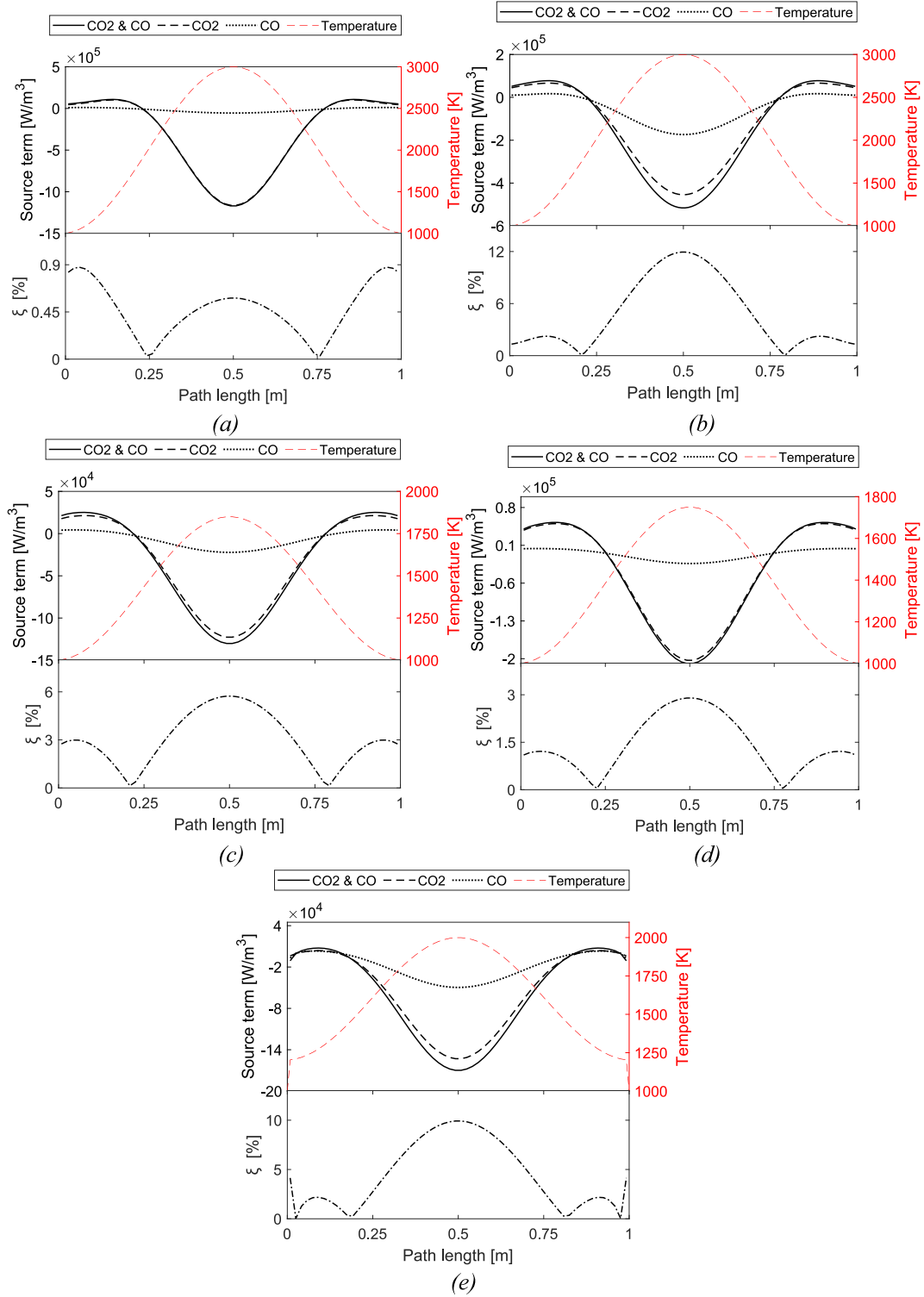


Fig. 4. Radiative source terms for conditions representing: (a) rich CO_2 conditions (Case 4); (b) rich CO conditions (Case 5); (c) propane-air combustion (Case 6); (d) propane-oxyfuel combustion (Case 7); and (e) a blast furnace (Case 8). The error introduced by treating CO as transparent are included beneath the respective source term profiles.

CO_2/CO ratio results in a negligible impact of CO on radiation across the domain. In contrast, under blast furnace conditions (Case 8), where the CO_2/CO ratio is low, Fig. 4e demonstrates a more-pronounced contribution of CO to the total radiation, again with the maximum effect located at the domain center. Overall, these results indicate that CO_2 dominates the radiative source term for the studied range of temperatures and CO_2/CO -mixtures. However, CO can contribute appreciably to

the total radiative emission under conditions characterized by low CO_2/CO -ratios.

For further examination of these trends, the radiation framework is employed to assess the impacts of CO and on the gas-mixture emissivity. Fig. 5a presents the CO error by Eq. (6) for temperatures and species fractions related to the rich CO_2/CO -mixtures of Cases 4 and 5, while Fig. 5b-e includes conditions associated with CO_2 -plumes, propane-air,

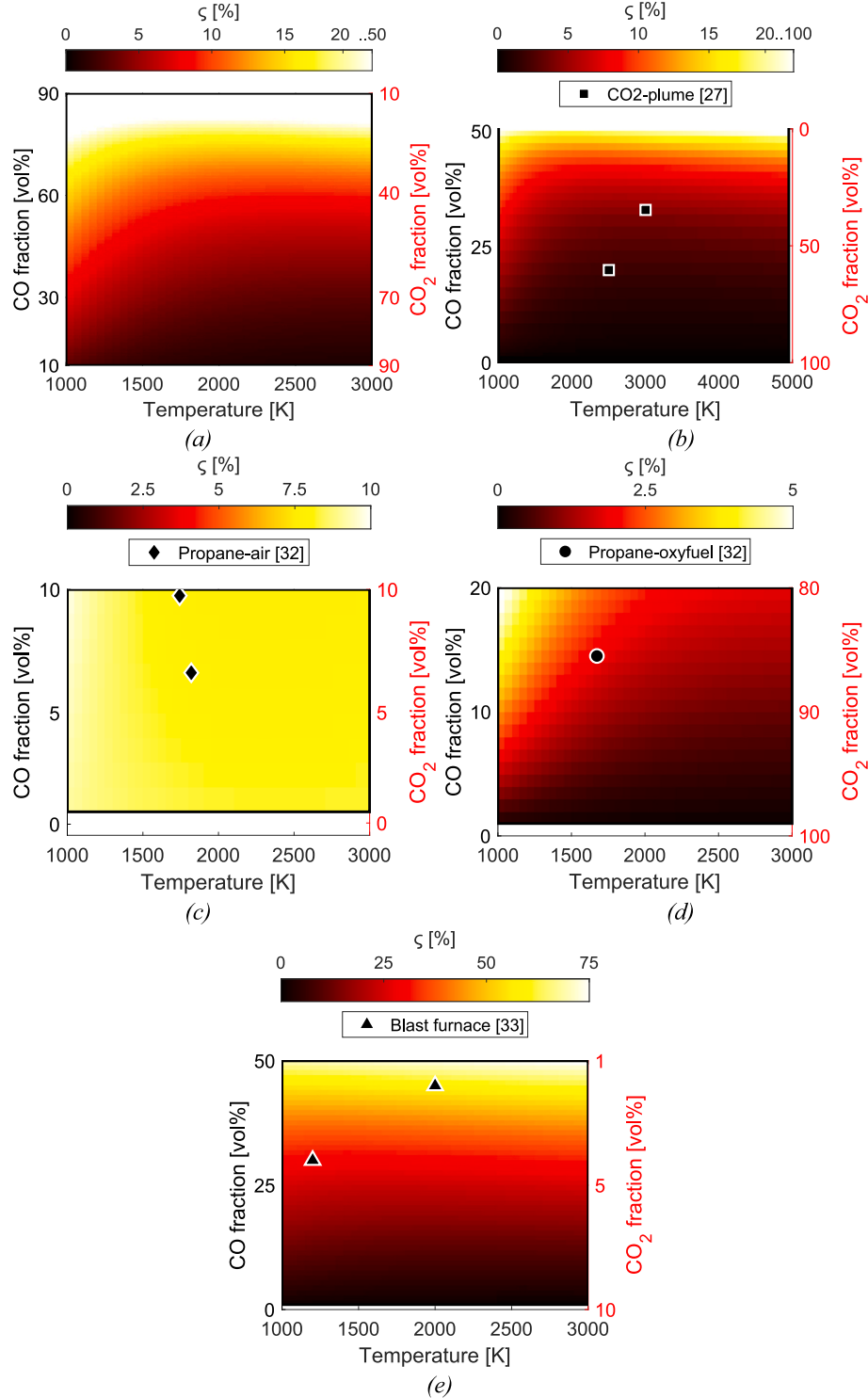


Fig. 5. The emissivity error for treating CO as transparent at five CO_2/CO /transparent gas mixtures, with CO_2 fractions spanning: a) 0.1–0.9 (no transparent gas); b) 0–1 (moderate transparent gas mixture); c) 0.01–0.1 (rich transparent gas mixture); d) 0.8–1 (no transparent gas); and e) 0.01–0.1 (moderate transparent gas mixture). The conditions similar to those reported in [27] [32] [33] is included in the relevant figures. The red y-axis denotes the CO_2 fraction. The pressure path length is 0.01 atm m in all cases. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

propane-oxyfuel, and blast furnace conditions (Cases 6–10). Accordingly, Fig. 5 spans five distinct CO_2/CO ratios, indicated by boxed regions, with the CO_2 fractions highlighted in red. Fig. 5a shows a weak impact of CO on the gas-mixture emissivity in the presence of lean CO fractions and high temperatures, whereas higher errors are achieved for richer CO fractions and lower temperatures. For CO_2/CO ratios related to the CO_2 -plume [27] (ratios above unity), Fig. 5b reveals an error within 5%. It is also observed that, within the investigated regime, although chemical equilibrium is not fully attained at 2100–2500 K, the associated uncertainty in the CO concentration resulting from CO_2 dissociation has a limited impact on the radiative conclusions. The error by Eq. (6) increases up to 20% with extremely low CO_2/CO ratios (~ 0.25), achievable at temperatures above 3800 K, which significantly dissociates CO_2 . For CO_2/CO ratios linked to propane-air and propane-oxyfuel combustion conditions, Fig. 5c-d, similar to Fig. 5a, indicates a limited influence of CO on the gas-mixture emissivity. However, CO exhibits a more-pronounced influence under conditions associated with lower temperatures (1000–2000 K) and under CO-rich and CO_2 -lean conditions, which are relevant to the blast furnace case. Further insight is obtained by evaluating the Planck distributions for both species at 1200 K and at a low CO_2/CO ratio of 0.21 (Fig. 6), representative of blast furnace conditions. Under these conditions, a broadening of the CO-band around 1900 cm^{-1} is observed, which significantly reduces the band overlap with CO_2 . Consequently, neglecting the CO contribution to the total gas-mixture emissivity leads to an underprediction by Eq. (6) of approximately 25%.

3.3. Radiation calculations based on CFD profiles

The local radiative contributions of CO_2 and CO under CFD-derived conditions are further assessed by predicting the radiative source term using the radiation framework. Case 9 is defined along a cross-section of the furnace diameter, with gas and wall temperatures and species compositions extracted from the CFD framework. The radial profiles at the location of peak temperature—approximately at the plasma torch outlet, where the CO concentration is highest—are provided in the *Supplementary Material* (Fig. S4).

The predicted radiative source terms for these radial profiles (Case 9) are shown in Fig. 7, along with the corresponding gas and wall temperatures. In contrast to the symmetric profiles observed for Cases 4–8, the distributions are asymmetric due to the tilting of the plasma torch, which produces a non-uniform radial temperature field. Near the left wall, the source term is slightly positive due to the temperature gradient between the wall ($\sim 1220\text{ K}$) and the gas ($\sim 1180\text{ K}$), which leads to absorption. Adjacent to the domain center, the source term becomes positive as the gas absorbs radiation from the peak temperature region, whereas at the domain center, the gas exhibits significant radiative emission. Close to the right wall, which is colder ($\sim 1180\text{ K}$) than the

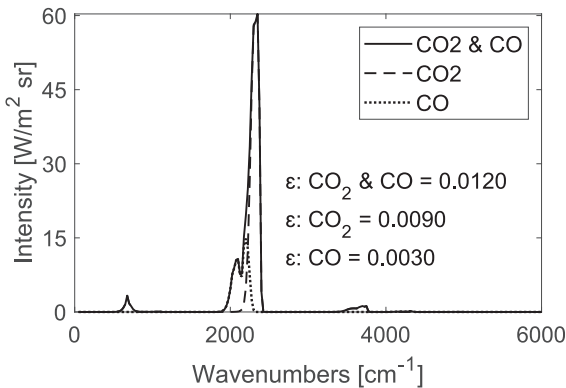


Fig. 6. Planck distributions at 1200 K along with a CO_2/CO ratio of 0.21. The pressure path length is 0.01 atm m.

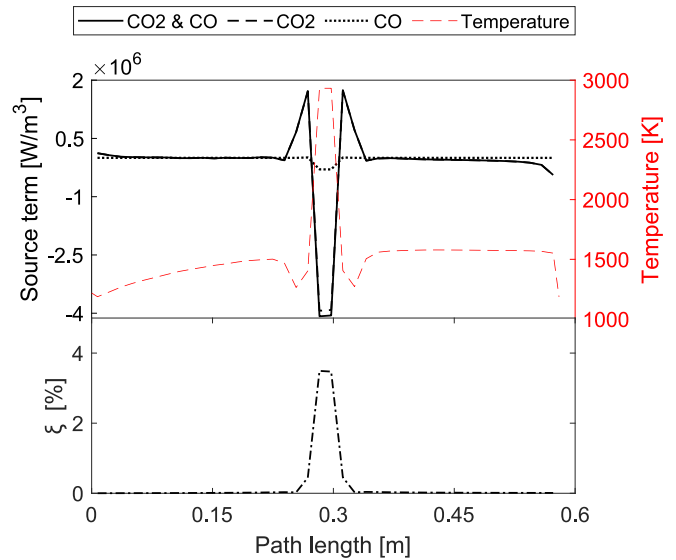


Fig. 7. Radiative source terms at the plasma torch outlet (for Case 9). The error introduced by treating CO as transparent is included beneath the source term profile.

adjacent gas ($\sim 1500\text{ K}$), the gas emits radiation, and the source term becomes negative. The results confirm that CO_2 dominates the radiative contribution, while CO primarily contributes to emission in the domain center, where its concentration is highest. Here, the peak error by Eq. (6) is less than 4%.

Evaluating the Planck distributions for both species under peak temperature (2932 K) conditions reveals a spectral overlap between CO and CO_2 in the waveband centered between 1900 and 2400 cm^{-1} (Fig. 8). This band overlap significantly attenuates the effective radiative contribution of CO, resulting in CO_2 dominating the total gas radiation within the furnace. The sensitivity of the selected pressure path length is shown in Fig. 8b-c, revealing, for increasing pressure path lengths, a progressively larger influence of CO in the infrared bands near the strong 2100 cm^{-1} band and the weaker 4200 cm^{-1} band. Saturation of the infrared bands of CO_2 for progressively larger pressure path lengths is also observed, as well as the overlap between CO and CO_2 near 2100 cm^{-1} . However, the effect on gas-mixture emissivity for treating CO as transparent remains within 5% due to the stronger emerging bands of CO_2 as seen in Fig. 8.

Under these conditions, the CO contribution to the total gas emissivity is, according to Eq. (6), $\sim 5\%$. This implies that if CO is considered a transparent gas in the radiative transfer sub-model within the CFD framework, the emissivity predicted for about 0.12% of the furnace gas volume within the CFD domain—which holds peak conditions where the CO fractions are equal to or greater than 30%—is accurate to within 95%. Furthermore, if the CO fractions were instead assigned to CO_2 , the resulting error would exceed that from treating CO as a transparent gas, because the artificially increased CO_2 concentration would enhance the gas's radiative contribution through more pronounced CO_2 infrared emission bands.

Case 10 extends this analysis by examining the radiative contributions along a line-of-sight path based on the CFD-simulated fields, originating from the plasma torch outlet, which extends 0.175 m into the furnace. The cumulative radiative intensity along the domain centerline is shown in Fig. 9. Across the entire optical path, CO_2 remains the dominant radiating species, while CO contributes only weakly to the total gas radiation, with its highest contribution being in the peak temperature zone. The corresponding mid-furnace CFD fields, illustrating the high-temperature CO_2 -plume exiting the plasma torch in the pilot rotary kiln, are provided in the *Supplementary Material* (Fig. S5).

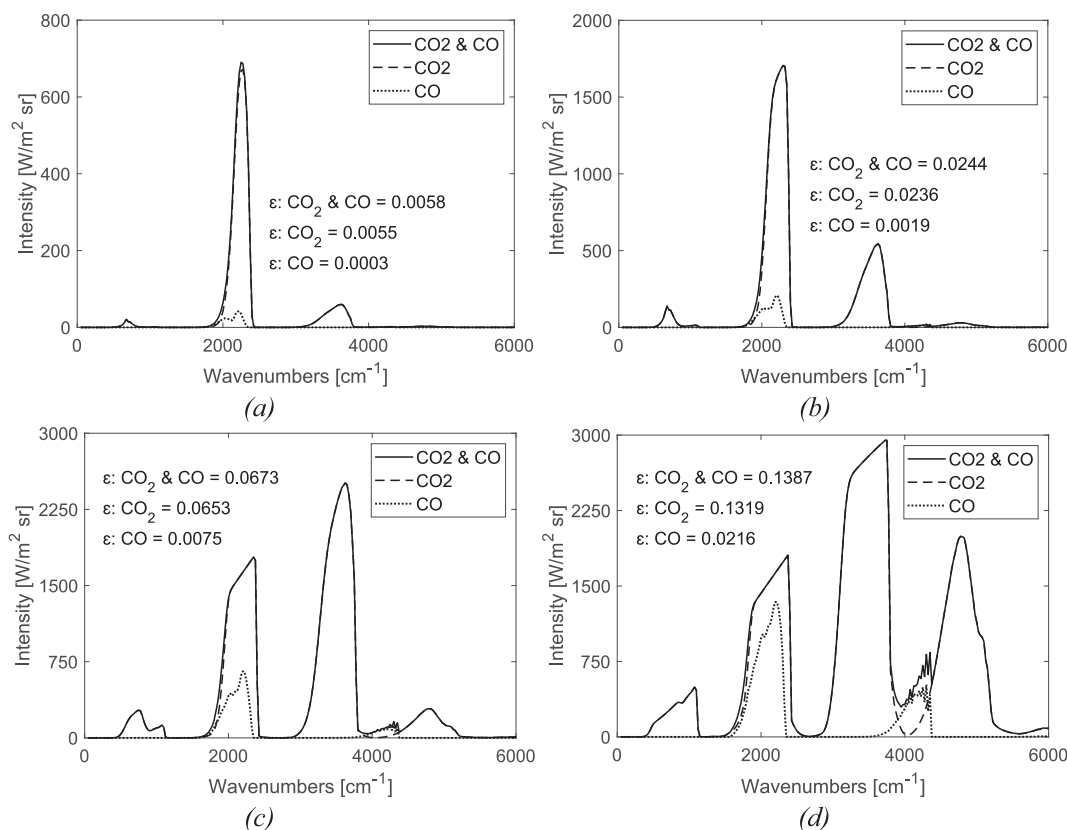


Fig. 8. Planck distributions under peak temperature conditions. The pressure path length spanning a) 0.01 atm m, b) 0.1 atm m, c) 1 atm m, and d) 10 atm m.

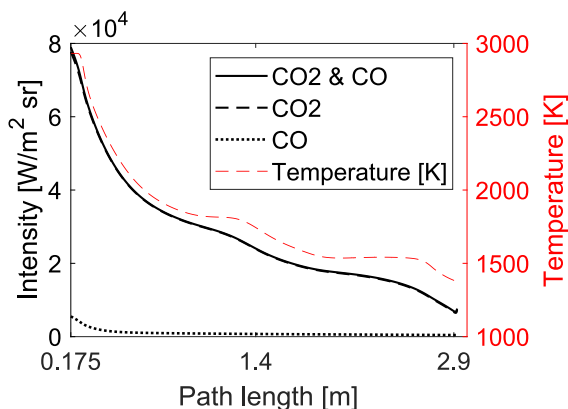


Fig. 9. Radiative intensities along the centerline of the CFD-simulated fields (for Case 10).

4. Conclusions

This work evaluates the radiative effects of CO in CO_2 -mixtures under conditions representative of high-temperature CO_2 -plumes. Two temperature zones are identified: zones of peak temperatures associated with CO_2 -dissociation, and colder zones influenced by CO oxidation. The analysis includes a chemistry framework that is used to predict the species composition within these zones, while a separate radiation modeling framework isolates and evaluates the individual radiative contributions of CO_2 and CO . In addition, the models are applied to a CFD model that incorporates chemistry, radiation, and turbulence sub-models to simulate the working gas plume of a thermal plasma-heated kiln, with data gathered from previous work.

The results show that, within the specific thermochemical and op-

tical regime representative of the investigated CO_2 -plasma-heated system, the timescales are sufficiently short for the chemical equilibrium assumption to be valid within the 3000 K temperature range. While the timescales are not necessarily sufficient to attain chemical equilibrium in the 2100–2500 K range, the relatively limited CO_2 dissociation in this range results in comparatively low CO concentrations. Further, the radiative contribution of CO can be neglected and treated as a transparent gas in gas radiation modeling with limited impact on accuracy. In particular, the results support this simplification for temperatures of approximately 1200–3000 K and CO/CO_2 ratios of unity or below. In these conditions, the error from neglecting CO is less than 6% in the peak radiative source term and about 7% in gas-mixture emissivity. Increasing radiative contributions from CO are only observed under two limiting conditions: for CO/CO_2 ratios substantially above unity, particularly under CO -rich conditions; and at temperatures above approximately 3800 K, where extensive CO_2 dissociation increases the CO concentrations while depleting CO_2 , resulting in a greater radiative impact from CO . This quantitative criterion provides a basis for simplifying the CO treatment in gas radiation models.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Klas Andersson reports financial support was provided by Swedish Energy Agency. Klas Andersson reports financial support was provided by European Union. Fredrik Normann reports financial support was provided by LKAB. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.applthermaleng.2026.133730>.

Data availability

No data was used for the research described in the article.

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