

FULL-SPECTRUM k -DISTRIBUTION CORRELATIONS FOR CARBON DIOXIDE MIXTURES

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ABSTRACT

Full-Spectrum k -distributions (or absorption-line blackbody distribution functions) have been calculated for CO₂ from the new HITEMP database for a large number of gas temperatures, and an approximate mathematical correlation is presented. This correlation supersedes the older one by Denison and Webb, which was based on the HITRAN92 database. Comparison between the correlations shows that, while giving excellent results for temperatures below 1000K, the older correlation seriously underpredicts radiative losses at high temperatures.

NOMENCLATURE

a = weight function for the FSCK method
 \bar{a} = weight function for the SLW model
 C_{lmn} = correlation coefficients
 f = k -distribution function, cm
 g = cumulative k -distribution
 I = radiative intensity, W/m²sr
 k = absorption coefficient variable, cm⁻¹
 k^* = correlated absorption coefficient, cm⁻¹
 \tilde{k} = absorption coefficient in the SLW model, cm⁻¹
 k_η = spectral absorption coefficient at reference state, cm⁻¹
 L = geometric length, m
 L_m = mean beam length, m
 p = pressure, bar
 P = nondimensional parameter used in the correlation
 q = radiative heat flux, W/m²
 s, s' = distance along path, m
 T = temperature, K
 u = scaling function for absorption coefficient
 V = volume
 x, \underline{x} = mole fraction

GREEK SYMBOLS

η = wavenumber, cm⁻¹
 ϵ = wall emittance
 ϕ = composition variable
 $\bar{\Phi}$ = scattering Phase function
 κ = absorption coefficient, cm⁻¹
 Ω = solid angle, sr
 σ_s = scattering coefficient, cm⁻¹

SUBSCRIPTS

0 = reference state
 b = blackbody emission
 g = gas
 i = range of the cumulative k -distribution in the SLW model
 P = Planck Mean
 w = wall
 η = spectral

INTRODUCTION

Radiative heat transfer in gases has important applications ranging from combustion systems to modeling atmospheric processes. The most accurate radiative heat transfer evaluations are line-by-line calculations, which are, however, very time intensive and require large computer resources. Therefore, many studies have been devoted to band models and global models. To date the most popular global model for nongray radiation calculation has been the weighted-sum-of-gray-gases approach, which was first presented by Hottel¹ within the framework of the zonal method. Modest^{2,3} has demonstrated that this approach can be applied to the directional equation of transfer and, therefore, to any solution method for the equation of transfer, provided the boundaries are black and the medium is nonscattering. In this method the nongray gas is replaced by a number of gray gases, for which the heat transfer rates are calculated independently. The total heat

flux is then found by adding the heat fluxes of the gray gases after multiplication with certain weight factors.

Today it is known that the method can also be applied to reflecting (albeit gray) walls and to gray scattering media, to variable absorption coefficients as long as they obey certain rules, and that accurate gray-gas coefficients can be obtained from high-resolution databases, mostly through the extensions to made by Denison and Webb^{4–8}. Their approach, which they call Spectral-Line-Based Weighted-Sum-of-Gray-Gases, or SLW, can finite-difference the spectral line structure of molecular gases to any desired accuracy. Their calculations show that extremely accurate results (compared to LBL benchmarks) can be obtained for homogeneous gas mixtures, using only three or four spectral calculations and – to a lesser extent – also in mixtures with varying temperatures and concentrations. A very similar method, the Absorption Distribution Function (ADF) model was developed by Rivière and coworkers^{9–11} and applied to one-dimensional mixtures of water vapor and carbon dioxide with various temperature and concentration profiles. They also extended the approach to include fictitious gases¹¹. Very recently, Modest and coworkers^{12,13} have developed the Full-Spectrum k -distribution (FSK) method and showed that the WSGG method, as applied in the SLW and ADF forms, is just a crude implementation of that more advanced method.

In a number of papers, Denison and Webb^{4–8,14,15} have also presented relatively simple correlations for the evaluation of the full-spectrum k -distributions (or absorption-line blackbody distribution function, as Denison and Webb call them). Unfortunately, these correlations are based on the outdated HITRAN92 database¹⁶ (together with some high-temperature extensions). Therefore, the total emissivities precalculated by these correlations generally are considerably lower than those from experiment, due to missing “hot lines” in the HITRAN database. It is the purpose of the present paper to provide a new full-spectrum k -distribution correlation, based on the HITEMP database¹⁷, for use with the SLW, ADF and FSK methods.

THEORETICAL BACKGROUND

Like the WSGG method the Full-Spectrum k -Distribution (FSK) demands that, except for the absorption coefficient, no other radiative property varies across the spectrum, and then attempts to integrate the radiative transfer equation across the entire spectrum before solving it. And, like narrow band k -distributions, this is achieved by reordering the absorption coefficient into a monotonically increasing function. However, in the full-spectrum case allowance must be made for a blackbody intensity (or Planck function) varying across the spectrum. This has been described in the original paper by Modest original paper by Modest and Zhang¹².

As with conventional band models, the treatment of nonhomogeneous media is more problematic, since it is not possible to

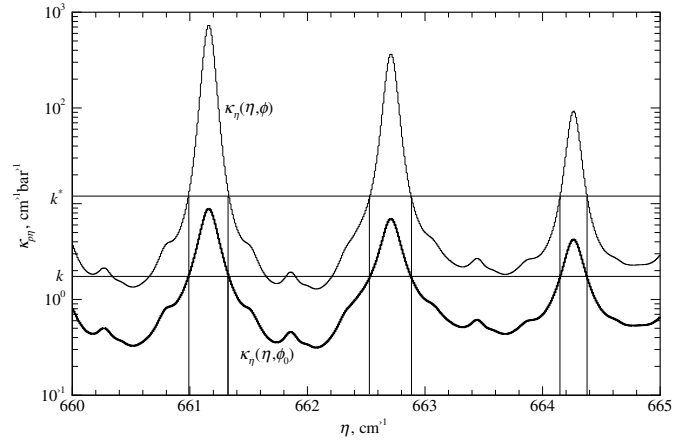


Figure 1. Extraction of k -distributions from spectral absorption coefficient data (thick line is for CO₂ in nitrogen, across a small portion of the 15 μ m band, $p = 1.0$ bar, $T = 296$ K; thin line is an artificially created correlated absorption coefficient).

develop exact k -distributions for arbitrary absorption coefficients in nonhomogeneous media. Thus, to develop a full-spectrum k -distribution method for nonhomogeneous media one must assume that the spectral absorption coefficient is correlated or even obeys the scaling approximation¹³: defining a vector $\underline{\phi}$ that contains the composition variables that affect the absorption coefficient, i.e., temperature T , pressure p , and mole fractions of gases (or the volume fraction of small particles, if present) \underline{x} , the absorption coefficient may be written as

$$\kappa_{\eta}(\eta, \underline{\phi}) = \begin{cases} k_{\eta}^*(\underline{\phi}, k_{\eta}), & \text{correlated,} \\ k_{\eta}(\eta) u(\underline{\phi}, \underline{\phi}_0), & \text{scaled,} \end{cases} \quad (1)$$

where $k_{\eta}(\eta) = \kappa_{\eta}(\eta, \underline{\phi}_0)$ is the absorption coefficient at a reference state $\underline{\phi}_0 = (T_0, p_0, \underline{x}_0)$. If the absorption coefficient is correlated then, at every wavenumber where $\kappa_{\eta}(\eta, \underline{\phi}_0)$ has one and the same value k , $\kappa_{\eta}(\eta, \underline{\phi})$ always also has one unique value $k^*(\underline{\phi}, k)$, as illustrated in Fig. 1 for a small part of a CO₂ band. If the ratio k_{η}^*/k_{η} is constant (not a function of k_{η}) then the absorption coefficient is scaled.

For a medium with such a correlated absorption coefficient, the RTE in the reordered absorption coefficient $k(g_0)$ becomes the Full-Spectrum Correlated k -distribution (FSCK) method¹³,

$$\frac{dI_g}{ds} = k^*(T_0, \underline{\phi}, g_0) \left[a(T, T_0, g_0) I_b(T) - I_g \right] - \sigma_s \left(I_g - \frac{1}{4\pi} \int_{4\pi} I_g(\hat{s}') \Phi(\hat{s}, \hat{s}') d\Omega' \right), \quad (2)$$

subject to the boundary condition

$$I_g = I_{wg} = \epsilon_w a(T_w, T, g_0) I_{bw} + (1 - \epsilon_w) \frac{1}{\pi} \int_{\hat{n} \cdot \hat{s} < 0} I_g |\hat{n} \cdot \hat{s}| d\Omega. \quad (3)$$

Here,

$$I_g = I_k / f(T_0, \underline{\phi}_0, k) = \int_0^\infty I_\eta \delta(k - \kappa_\eta(\eta, \underline{\phi}_0)) d\eta \Big| f(T_0, \underline{\phi}_0, k), \quad (4)$$

$$g_0(T_0, \underline{\phi}_0, k) = \int_0^k f(T_0, \underline{\phi}_0, k) dk, \quad (5)$$

$$a(T, T_0, g_0) = \frac{f(T, \underline{\phi}_0, k)}{f(T_0, \underline{\phi}_0, k)} = \frac{dg(T, \underline{\phi}_0, k)}{dg_0(T_0, \underline{\phi}_0, k)}. \quad (6)$$

and the total intensity is evaluated from

$$I = \int_0^\infty I_\eta d\eta = \int_0^\infty I_k dk = \int_0^1 I_g dg_0.$$

In this set of equations, the ‘‘spectral variable’’ g_0 is a Planck function weighted cumulative k -distribution, evaluated from the Planck function weighted full-spectrum k -distribution, defined as

$$f(T, \underline{\phi}_0, k) = \frac{1}{I_b(T)} \int_0^\infty I_{b\eta}(T) \delta(k - \kappa_\eta(\eta, \underline{\phi}_0)) d\eta. \quad (7)$$

Equation (2) requires the choice of a reference condition at which the exact absorption coefficient is used (and is simply assumed to be ‘‘correlated’’ with that state at the local conditions inside the medium), and a reference temperature, T_0 , at which the cumulative k -distribution is evaluated. The weight function a represents a stretching function between the local spectral variable g , and the reference temperature value g_0 . The Full-Spectrum Correlated k -distribution method of equation (2-7) requires two sets of full-spectrum k -distributions $k(T_0, \underline{\phi}, g_0)$ (for the evaluation of the absorption coefficient in equation (2); absorption coefficient evaluated at local conditions, Planck function at reference temperature T_0) and $k(T, \underline{\phi}, g)$ (for the evaluation of the weight function a ; absorption coefficient determined at reference condition, Planck function evaluated at local temperature). The availability of Denison and Webb’s correlation, or the new one presented in this paper, makes high-accuracy radiative heat transfer calculations possible without the need for lengthy pre-calculations from a spectral database.

If the absorption coefficient is scaled (as well as correlated), equation (2) reduces to the Full-Spectrum Scaled k -distribution

(FSSK) method,

$$\frac{dI_g}{ds} = ku \left[a(T, T_0, g_0) I_b(T) - I_g \right] - \sigma_s \left(I_g - \frac{1}{4\pi} \int_{4\pi} I_g(\hat{s}') \Phi(\hat{s}, \hat{s}') d\Omega' \right). \quad (8)$$

Whether the assumption of a correlated absorption coefficient is to be used (FSSK), or whether the absorption coefficient is to be scaled (FSSK), the exact k vs. g behavior can be employed only for a single reference state $\underline{\phi}_0$. Therefore, the choice of $\underline{\phi}_0$ is very important and should be optimized for any given problem (on the other hand, the reference Planck function temperature is only a mathematical convenience, and its choice does not affect the accuracy of calculations). Modest and Zhang¹² suggest, for a medium at constant total pressure p ,

$$\underline{x}_0 = \frac{1}{V} \int_V \underline{x} dV, \quad (9)$$

$$\kappa_P(T_0, \underline{x}_0) = \frac{1}{V} \int_V \kappa_P(T, \underline{x}) I_b(T) dV, \quad (10)$$

i.e., volume-averaged mole fraction and a Planck-mean temperature based on average emission from the volume.

In the correlated- k method, the $k(T, \underline{\phi}_0, g)$ are then determined, followed by evaluation of $k^* = k(T_0, \underline{\phi}, g_0)$ making the assumption of corresponding g -values (and its resulting errors). If a scaled absorption coefficient is to be used, scaling functions must be found, and Modest and Zhang¹² suggest the implicit relation

$$\int_0^\infty I_{b\eta}(T_0) \exp(-\kappa_\eta(\eta, \underline{\phi}) L_m) d\eta = \int_0^\infty I_{b\eta}(T_0) \exp[-\kappa_\eta(\eta, \underline{\phi}_0) u(\underline{\phi}, \underline{\phi}_0) L_m] d\eta, \quad (11)$$

i.e., forcing correct evaluation of radiation leaving from a homogeneous slab equal in width to the mean beam length, L_m . Using k -distributions this becomes

$$\int_0^1 \exp[-k^*(T_0, \underline{\phi}, g) L_m] dg = \int_0^1 \exp[-k(T_0, \underline{\phi}_0, g_0) u(\underline{\phi}, \underline{\phi}_0) L_m] dg_0. \quad (12)$$

Both methods are about equally efficient numerically: besides the evaluation of $k(T, \underline{\phi}_0, g)$ [needed for both methods to evaluate $k(g_0)$ and the weight function a], for a correlated absorption coefficient, k -distributions must be evaluated for all states

ϕ (with a Planck function based on the reference temperature). For a scaled absorption coefficient, the same k -distributions are needed, but here for the evaluation of the scaling functions u . However, the scaled- k method holds two advantages over its correlated cousin: (i) for a poorly correlated absorption coefficient the scaling function from Eq. (12) can partially correct for this lack of correlation, and (ii) alternatively, a simplified scaling function can be chosen without the use of Eq. (12), thus reducing the number of cumbersome k -distribution evaluations.

Both the FSCK and FSSK methods are “exact” within the restrictions of an absorption coefficient that is correlated or scaled, as well as gray walls and gray scattering properties.

SLW Method If equation (2) is integrated using a crude trapezoidal scheme, i.e., the spectrally-varying absorption coefficient $k(g_0)$ is replaced by a single, constant value for the i -th finite range of g_0 spanning across $g_{0,i-1} < g_0 < g_{0,i}$, then the RTE reduces to the SLW method⁴⁻⁸,

$$\frac{dI_i}{ds} = \bar{k}_i(T_0, \underline{\phi}) (\bar{a}_i(T, T_0) I_b(T) - I_i) - \sigma_s \left(I_i - \frac{1}{4\pi} \int_{4\pi} I_i(\hat{s}') \Phi(\hat{s}, \hat{s}') d\Omega' \right), \quad i = 1, \dots, N, \quad (13)$$

subject to the boundary condition

$$I_{wi} = \epsilon_w \bar{a}_i(T_w, T_0) I_b(T_w) + (1 - \epsilon_w) \frac{1}{\pi} \int_{\hat{n} \cdot \hat{s} < 0} I_i |\hat{n} \cdot \hat{s}| d\Omega, \quad (14)$$

where

$$I_i = I_g(\bar{k}_i)(g_{0,i} - g_{0,i-1}). \quad (15)$$

The weight function \bar{a}_i is evaluated from Eq. (6) as

$$\begin{aligned} \bar{a}_i(T, \underline{\phi}_0) &= \int_{g_{0,i-1}}^{g_{0,i}} a(T, T_0, g_0) dg_0(T_0, \underline{\phi}_0) \\ &= \int_{g_{i-1}}^{g_i} dg(T, \underline{\phi}_0) = g_i(T, \underline{\phi}_0) - g_{i-1}(T, \underline{\phi}_0). \end{aligned} \quad (16)$$

Thus, \bar{a}_i is the i -th finite range of the cumulative k -distribution evaluated at the local Planck function temperature. The \bar{k}_i is an average value of $k^*(T_0, \underline{\phi}, g_0)$ over the range $g_{0,i-1} < g_0 \leq g_{0,i}$ (i.e., the value from the k -distribution evaluated from the local absorption coefficient and the Planck function evaluated at the reference temperature), and is calculated as^{4,6,7}

$$\bar{k}_i(T_0, \underline{\phi}) = \sqrt{k_{i-1}(T_0, \underline{\phi}) k_i(T_0, \underline{\phi})}. \quad (17)$$

These rather complicated relationships for \bar{a} and k^* were correctly deduced by Denison and Webb^{6,7}, well before a solid theoretical foundation describing the interrelationships between k -distributions was developed by Modest¹³.

FULL-SPECTRUM k -DISTRIBUTION CORRELATION

In our present work a new correlation for the cumulative full-spectrum k -distribution of carbon dioxide was developed, based on the latest spectral database, HITEMP¹⁷. A relationship similar to the original one by Denison and Webb, using a hyperbolic tangent function, was used here, resulting in

$$g = \frac{1}{2} \tanh [P(T_P, T_g, k)] + \frac{1}{2}, \quad (18)$$

and

$$P(T_P, T_g, k) = \sum_{l=0}^3 \sum_{m=0}^3 \sum_{n=0}^3 C_{lmn} \left(\frac{T_P}{2500 \text{ K}} \right)^n \left(\frac{T_g}{2500 \text{ K}} \right)^m (\log_{10} k)^l, \quad (19)$$

where k is in cm^{-1} . The C_{lmn} are the correlation coefficients and are listed in Table 1. Some representative results of the correlation are shown in Figs. 2 and 3 and are compared with “exact” HITEMP results and results from Denison and Webb’s correlation. Figure 2 shows the full-spectrum k -distribution $k(T_P = 1000\text{K}, T_g, g)$, with the absorption coefficient evaluated at a number of different temperatures, each time for the same Planck function temperature $T_P = 1000\text{K}$. The Denison and Webb correlation is expected to do well for relatively low T_g , since the augmented HITRAN database should be fairly reliable up to about 1000K. This was found to be true, although the Denison and Webb correlation is not very good for very small k values. The Denison and Webb correlation starts showing inaccuracies for $T_g = 1500\text{K}$, and hardly changes any more if the gas temperature is raised to 2500K. In Fig. 3 the gas temperature (at which the absorption coefficient is evaluated) is fixed at $T_g = 2000\text{K}$ and, as expected, the Denison and Webb correlation emphasizes smaller values of k (larger values of g for same k) due to missing hot lines, and this occurs regardless of Planck function temperature.

The cumulative k -distributions function correlations may also be used to calculate the total emissivity of isothermal CO_2 - N_2 mixtures, in order to compare them with experimental data or correlations based on such data. This is done in Fig. 4, comparing total emissivities obtained from the present correlations with those obtained from Denison and Webb’s correlation, and those calculated from Leckner’s model¹⁸, which is based on (fairly dated) experimental data. It is observed that, as expected, the Denison and Webb underpredicts high-temperature emissivities due to missing hot lines, particularly for intermediate optical thickness $p_a L$, p_a being the partial pressure of CO_2 , and L

Table 1. Coefficients for the full-spectrum k -distribution of carbon dioxide.

	l	$m \setminus n$	0	1	2	3
C_{lmn}	0	0	1.33674	5.25708	-3.24722	0.46505
		1	1.23941	-31.45171	20.99698	-4.74781
		2	-0.79347	51.68059	-29.28778	3.25643
		3	0.59501	-26.08660	12.44131	0.27330
	1	0	-0.43810	16.95696	-20.09186	6.30367
		1	4.97136	-93.82591	1.11843E+02	-33.49779
		2	-7.68786	1.49486E+02	-1.63980E+02	40.92052
		3	3.80727	-74.92827	76.91119	-16.15887
	2	0	-0.69538	11.15900	-13.87794	3.78667
		1	4.86503	-61.32442	70.20234	-15.10310
		2	-8.22873	98.89924	-1.04446E+02	16.90963
		3	4.24771	-49.93400	49.71699	-6.01034
3	0	-0.21523	2.52271	-3.16365	0.81276	
	1	1.45256	-13.94386	16.13794	-3.34811	
	2	-2.46793	22.52534	-24.47289	4.05981	
	3	1.27270	-11.34729	11.76446	-1.56612	

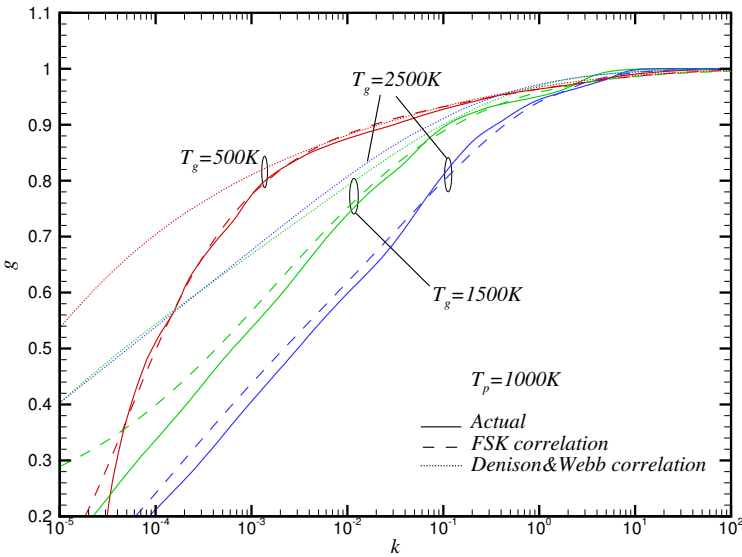


Figure 2. Actual and approximate full-spectrum k -distribution function for CO_2 calculated at a fixed Planck function temperature of 1000K with absorption coefficient taken at various temperatures.

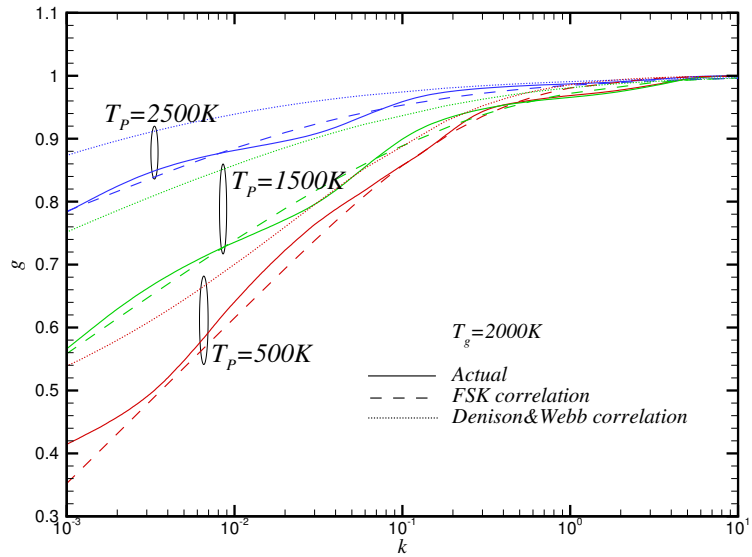


Figure 3. Actual and approximate full-spectrum k -distribution function for CO_2 calculated at a fixed gas temperature of 2000K with Planck function evaluated at various temperatures.

the column width. However, it is also seen that HITEMP (and the present correlation) overpredict emissivities as compared to Leckner's model. As indicated by Modest and Bharadwaj¹⁹,

HITEMP displays some questionable behavior in the band wings of CO_2 at temperatures above 1500K, which could explain these trends and which need to be resolved.

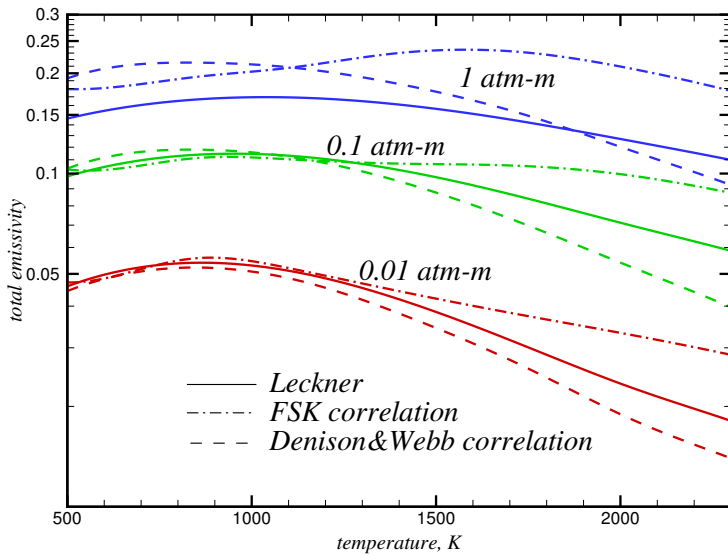


Figure 4. Total emissivities of CO₂ from Leckner, FSK correlation and Denison and Webb's correlation.

SAMPLE CALCULATIONS

A number of relatively simple one-dimensional examples considering CO₂-N₂ mixtures confined between two infinite parallel walls are presented to test the correlations. The $P-1$ approximation is employed here as the RTE solver since it is a very popular method with reasonable levels of effort and accuracy. In all examples the FSSK method has been employed for the spectral model (using HITEMP directly, using the present correlation, and using the Denison and Webb correlation). While the FSSK method is the most accurate method, the FSK and SLW methods could have been employed just as well. First an isothermal medium confined between two parallel, cold and black plates is considered. The medium is a nitrogen-carbon-dioxide mixture at 1500K, 1bar, with a 10% mole fraction of CO₂. Benchmark line-by-line results for the local heat flux are shown in Fig. 5, together with FSK result and those from correlations. It can be seen that the FSK result and the HITEMP correlation essentially coincide with the LBL results, while the Denison and Webb correlation shows a maximum error of around 20% due to missing "hot lines".

An extreme case of an isothermal hot layer adjacent to an isothermal cold layer is then taken to further test the correlations. The pressure and CO₂ mole fraction are constant throughout at 1bar and 10% respectively. The hot layer is at $T=2000\text{K}$ and has a fixed width of 50cm, while the cold layer is at 300K, and is of varying width. With FSK results and the HITEMP correlation being within 10% of the LBL result, Denison and Webb correlation has a maximum error of 60%. Many "hot lines" have waken up at this high temperature of 2000K, which have not been con-

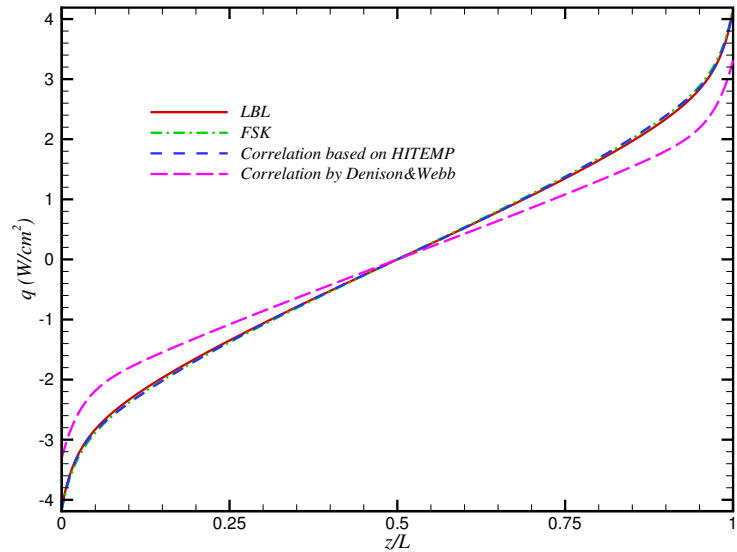


Figure 5. Local radiative flux in an isothermal N₂-CO₂ mixture ($T=1500\text{K}$, $p=1\text{bar}$, $x_{\text{CO}_2}=0.1$, $L=1\text{m}$) bounded by cold, black walls

sidered in Denison and Webb correlation.

The previous example with a step change in temperature were designed to be a worst-case scenario. As a last example we will consider nitrogen-carbon dioxide mixtures with smoothly varying temperature and mole fraction profiles, such as one may expect to occur in actual combustion applications. Figure 7 shows the radiative heat flux in a 1m thick gas layer with parabolic temperature and mole fraction profiles. The black walls are kept at 300K. In this problem the direct FSSK shows a maximum error of about 10% and, due to compensating errors, the present approximate k -distributions reduce this error to about 5%. The Denison and Webb correlation, while also benefiting from this compensating error, overshoots the LBL results by a maximum of about 15%.

SUMMARY AND CONCLUSIONS

An approximate correlation for full-spectrum k -distributions based on the HITEMP database was developed and tested through a number of one-dimensional problems. It was shown that this correlation can essentially achieve the same accuracy as full-spectrum k -distributions, directly calculated from the HITEMP database. The new correlation was also compared with that of Denison and Webb, which is based on the HITRAN92 database. It was found that the Denison and Webb correlation severely underpredicts radiation at high temperatures due to missing "hot lines".

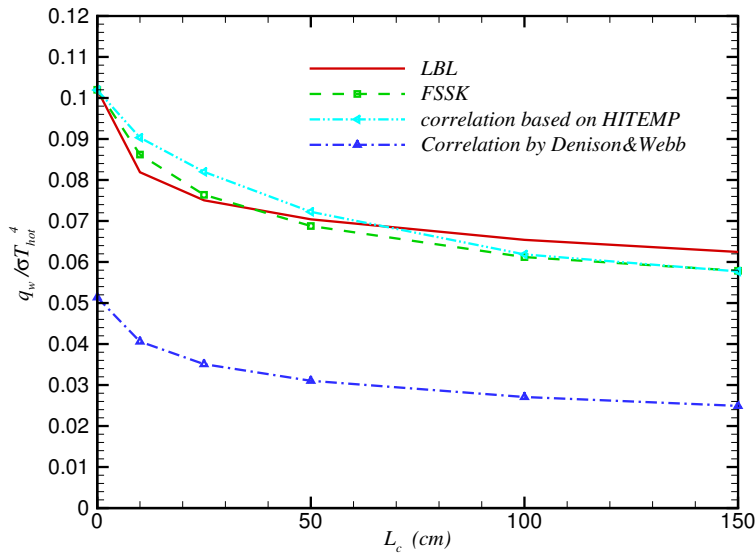


Figure 6. Radiative flux exiting from the cold column of a two-column CO₂-nitrogen mixture at different temperatures ($T_h = 2000\text{K}$, $L_h = 50\text{cm}$; $T_c = 300\text{K}$, L_c variable; uniform $p = 1\text{bar}$, $x_{\text{CO}_2} = 0.1$, cold and black walls on both sides).

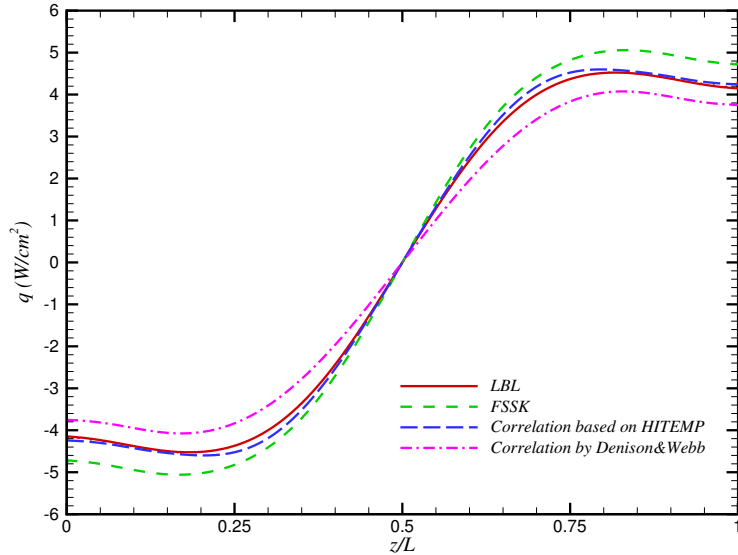


Figure 7. Radiative heat flux in a $L=1\text{m}$ thick CO₂-N₂ mixture with parabolic temperature and mole fraction profiles [$T = T_c - (T_c - T_w)(2z/L - 1)^2$, $x_{\text{CO}_2} = x_c - (x_c - x_w)(2z/L - 1)^2$, $T_c = 1750\text{K}$, $T_w = 300\text{K}$, $x_c = 0.2$, $x_w = 0.01$.]

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