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Full-Spectrum *k*-Distribution Correlations for Carbon Dioxide Mixtures

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Full-spectrum *k*-distributions (or absorption-line blackbody distribution functions) have been calculated for CO_2 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, although giving excellent results for temperatures below 1000 K, the older correlation seriously underpredicts radiative losses at high temperatures.

Nomenclature

a	=	weight function for the full-spectrum correlated k-distribution method
$\bar{a}$	=	weight function for the spectral-line-based weighted sum of gray gases (SLW) model
C_{lmn}	=	correlation coefficients
f	=	k-distribution function, cm
g	=	cumulative k-distribution
I	=	radiative intensity, W/m^2 sr
k	=	absorption coefficient variable, cm^{-1}
k_η	=	spectral absorption coefficient at reference state, cm^{-1}
k^*	=	correlated absorption coefficient, cm^{-1}
$\tilde{k}$	=	absorption coefficient in the SLW model, cm^{-1}
L	=	geometric length, m
L_m	=	mean beam length, m
P	=	nondimensional parameter used in the correlation
p	=	pressure, bar
q	=	radiative heat flux, W/m^2
s, s'	=	distance along path, m
T	=	temperature, K
u	=	scaling function for absorption coefficient
V	=	volume
$x, \boldsymbol{x}$	=	mole fraction
ϵ	=	wall emittance
η	=	wave number, cm^{-1}
κ	=	absorption coefficient, cm^{-1}
σ_s	=	scattering coefficient, cm^{-1}
Φ	=	scattering phase function
ϕ	=	composition variable
Ω	=	solid angle, sr

Subscripts

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

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I. 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 (LBL) 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 Hottel and Sarofim[1] 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 that 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, and to media with variable absorption coefficients, as long as they obey certain rules. Accurate gray-gas coefficients can be obtained from high-resolution databases, mostly through the extensions made by Denison and Webb.[4–8] Their approach, which they call spectral-line-based weighted sum of gray gases (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 et al.[9] and Pierrot et al.[10,11] 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 gases.[11] Very recently, Modest and Zhang[12] and Modest[13] have developed the full-spectrum k-distribution (FSK) method and showed that the weighted sum of gray gases (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 FSK (or absorption-line blackbody distribution function, as Denison and Webb call them). Unfortunately, these correlations are based

on the outdated HITRAN92 database,[16] together with some high-temperature extensions. Therefore, the total emissivities precalculated by these correlations are generally 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 FSK correlation, based on the HITEMP database,[17] for use with the SLW, ADF, and FSK methods.

II. Theoretical Background

Like the WSGG method, the FSK model demands that, except for the absorption coefficient, no other radiative property varies across the spectrum. It then attempts to integrate the radiative transfer equation (RTE) across the entire spectrum before solving it. 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 and Zhang.[12]

As with conventional band models, the treatment of nonhomogeneous media is more problematic because it is not possible to develop exact k-distributions for arbitrary absorption coefficients in nonhomogeneous media. Thus, to develop an FSK method for nonhomogeneous media, one must assume that the spectral absorption coefficient is correlated or even obeys the scaling approximation.[13] Given a vector $\boldsymbol{\phi}$ that contains the composition variables that affect the absorption coefficient [i.e., T, p, and mole fractions of gases (or the volume fraction of small particles, if present) $\boldsymbol{x}$], the absorption coefficient may be written as

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

where $k_\eta(\eta) = \kappa_\eta(\eta, \boldsymbol{\phi}_0)$ is the absorption coefficient at a reference state $\boldsymbol{\phi}_0 = (T_0, p_0, \boldsymbol{x}_0)$. If the absorption coefficient is correlated, then at every wave number where $\kappa_\eta(\eta, \boldsymbol{\phi}_0)$ has one and the same value k, then $\kappa_\eta(\eta, \boldsymbol{\phi})$ always has one unique value $k^*(\boldsymbol{\phi}, k)$, as illustrated in Fig. 1 for a small part of a CO_2 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) method,[13]

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

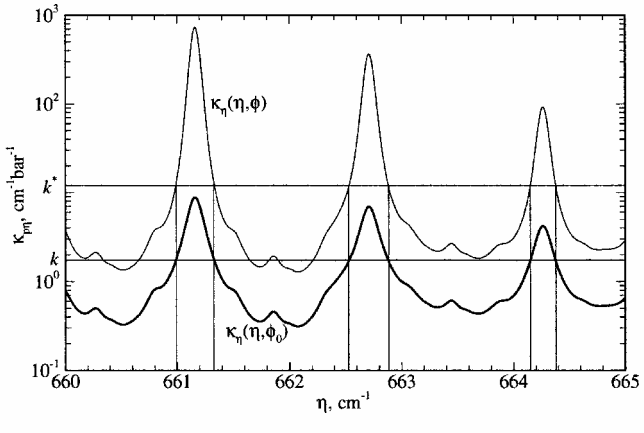

Fig. 1 Extraction of k-distributions from spectral absorption coefficient data: ——, CO_2 in nitrogen, across a small portion of the 15-μm band, $p = 1.0$ bar, and $T = 296$ K and ——, artificially created correlated absorption coefficient.

subject to the boundary condition

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

Here,

$$I_g = \frac{I_k}{f(T_0, \boldsymbol{\phi}_0, k)} = \frac{\int_0^\infty I_\eta \delta[k - \kappa_\eta(\eta, \boldsymbol{\phi}_0)]\, \mathrm{d}\eta}{f(T_0, \boldsymbol{\phi}_0, k)} \tag{4}$$

$$g_0(T_0, \boldsymbol{\phi}_0, k) = \int_0^k f(T_0, \boldsymbol{\phi}_0, k)\, \mathrm{d}k \tag{5}$$

$$a(T, T_0, g_0) = \frac{f(T, \boldsymbol{\phi}_0, k)}{f(T_0, \boldsymbol{\phi}_0, k)} = \frac{\mathrm{d}g(T, \boldsymbol{\phi}_0, k)}{\mathrm{d}g_0(T_0, \boldsymbol{\phi}_0, k)} \tag{6}$$

and the total intensity is evaluated from

$$I = \int_0^\infty I_\eta\, \mathrm{d}\eta = \int_0^\infty I_k\, \mathrm{d}k = \int_0^1 I_g\, \mathrm{d}g_0$$

In this set of equations, the "spectral variable" g_0 is a Planck function weighted cumulative k-distribution, evaluated from the Planck function weighted FSK defined as

$$f(T, \boldsymbol{\phi}_0, k) = \frac{1}{I_b(T)} \int_0^\infty I_{b\eta}(T) \delta[k - \kappa_\eta(\eta, \boldsymbol{\phi}_0)]\, \mathrm{d}\eta \tag{7}$$

Equation (2) requires the choice of a reference conditions 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), as well as 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 FSCK method of Eqs. (2–7) requires two sets of FSK $k(T_0, \boldsymbol{\phi}, g_0)$ [for the evaluation of the absorption coefficient in Eq. (2), the absorption coefficient evaluated at local conditions, and the Planck function at reference temperature T_0] and $k(T, \phi_0, g)$ (for the evaluation of the weight function a, the absorption coefficient determined at reference condition, and the 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 precalculations from a spectral database.

If the absorption coefficient is scaled (as well as correlated), Eq. (2) reduces to the full-spectrum scaled k-distribution (FSSK) method,

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

Whether the assumption of a correlated absorption coefficient is to be used (FSCK), 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 $\boldsymbol{\phi}_0$. Therefore, the choice of $\boldsymbol{\phi}_0$ is very important and should be optimized for any given problem, as indicated by Modest and Zhang.[12] (On the other hand, the reference Planck function temperature is only a mathematical convenience, and its choice does not affect the accuracy of calculations.)

Both methods are about equally efficient numerically: Besides the evaluation of $k(T, \boldsymbol{\phi}_0, g)$ [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 $\boldsymbol{\phi}$ (with a Planck function based on the reference temperature). For a scaled absorption coefficient, the same k-distributions are needed, but, in this case, for the evaluation of the scaling functions u (Ref. 12). However, the scaled-k method holds two advantages over the correlated version: 1) For a poorly correlated absorption coefficient,

the scaling function can partially correct for this lack of correlation. 2) Alternatively, a simplified scaling function can be chosen, 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 within those for gray walls and gray scattering properties.

SLW Method: If Eq. (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 ith finite range of g_0 spanning across $g_{0,i-1} < g_0 < g_{0,i}$], then the RTE reduces to the SLW method[4,8]

$$\frac{\mathrm{d}I_i}{\mathrm{d}s} = \tilde{k}_i(T_0, \boldsymbol{\phi})[\bar{a}_i(T, T_0)I_b(T) - I_i] - \sigma_s\left[I_i - \frac{1}{4\pi}\int_{4\pi} I_i(\hat{\boldsymbol{s}}')\Phi(\hat{\boldsymbol{s}}, \hat{\boldsymbol{s}}')\,\mathrm{d}\Omega'\right], \qquad i = 1, \ldots, N \quad (9)$$

subject to the boundary condition

$$I_{wi} = \epsilon_w \bar{a}_i(T_w, T_0)I_b(T_w) + (1-\epsilon_w)\frac{1}{\pi}\int_{\hat{n}\cdot\hat{s}<0} I_i|\hat{\boldsymbol{n}}\cdot\hat{\boldsymbol{s}}|\,\mathrm{d}\Omega \quad (10)$$

where

$$I_i = I_g(\tilde{k}_i)(g_{0,i} - g_{0,i-1}) \quad (11)$$

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

$$\bar{a}_i(T, \boldsymbol{\phi}_0) = \int_{g_{0,i-1}}^{g_{0,i}} a(T, T_0, g_0)\,\mathrm{d}g_0(T_0, \boldsymbol{\phi}_0) = \int_{g_{i-1}}^{g_i} \mathrm{d}g(T, \boldsymbol{\phi}_0) = g_i(T, \boldsymbol{\phi}_0) - g_{i-1}(T, \boldsymbol{\phi}_0) \quad (12)$$

Thus, $\bar{a}_i$ is the ith finite range of the cumulative k-distribution evaluated at the local Planck function temperature. The $\tilde{k}_i$ is an average value of $k^*(T_0, \phi, g_0)$ over the range $g_{0,i-1} < g_0 \le g_{0,i}$, that is, the value from the k-distribution evaluated with the local absorption coefficient and the Planck function evaluated at the reference temperature.[4,6,7] These rather complicated relationships for $\bar{a}$ and k^* were correctly deduced by Denison and Webb well before a solid theoretical foundation describing the interrelationships between k-distributions was developed by Modest.[13]

III. FSK Correlation

In our present work, a new correlation for the cumulative FSK of carbon dioxide was developed, based on the latest spectral database, HITEMP.[17] A relationship similar to the original one by Denison and Webb, using a hyperbolic tangent function, was used here, resulting in

$$g = \frac{1}{2}\tanh[P(T_P, T_g, k)] + \frac{1}{2} \quad (13)$$

$$P(T_P, T_g, k) = \sum_{l=0}^{3}\sum_{m=0}^{3}\sum_{n=0}^{3} C_{lmn}\left(\frac{T_P}{2500\ \mathrm{K}}\right)^n\left(\frac{T_g}{2500\ \mathrm{K}}\right)^m(\log_{10}k)^l \quad (14)$$

where k is per centimeter. 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[7] correlation. Figure 2 shows the FSK $k(T_P = 1000\ \mathrm{K}, T_g, g)$, with the absorption coefficient evaluated at a number of different temperatures, each time for the same Planck function temperature $T_P = 1000$ K. The Denison and Webb correlation is expected to do well for relatively low T_g because the augmented HITRAN database should be fairly reliable up to about 1000 K. 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[7] correlation starts showing inaccuracies for $T_g = 1500$ K and hardly changes if the gas temperature is raised to 2500 K. In Fig. 3, the gas temperature (at which the absorption coefficient is evaluated) is fixed at $T_g = 2000$ 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. 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, to compare them with experimental data or correlations

Table 1 Coefficients for the FSK of carbon dioxide

m/n	0	1	2	3
		$C_{lmn}, l=0$		
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
		$C_{lmn}, l=1$		
0	−0.43810	16.95696	−20.09186	6.30367
1	4.97136	−93.82591	111.843	−33.49779
2	−7.68786	149.486	−163.980	40.92052
3	3.80727	−74.92827	76.91119	−16.15887
		$C_{lmn}, l=2$		
0	−0.69538	11.15900	−13.87794	3.78667
1	4.86503	−61.32442	70.20234	−15.10310
2	−8.22873	98.89924	−104.446	16.90963
3	4.24771	−49.93400	49.71699	−6.01034
		$C_{lmn}, l=3$		
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

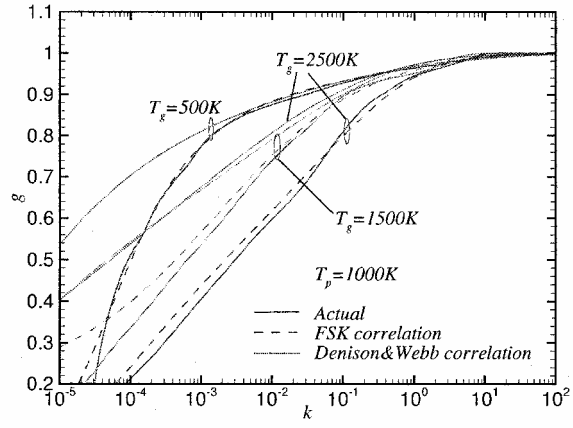

Fig. 2 Actual and approximate FSK function for CO_2 calculated at a fixed Planck function temperature of 1000 K with absorption coefficient taken at various temperatures.

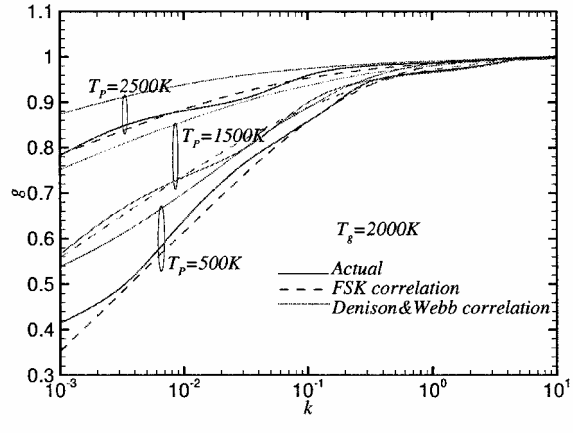

Fig. 3 Actual and approximate FSK for CO_2 calculated at a fixed gas temperature of 2000 K with Planck function evaluated at various temperatures.

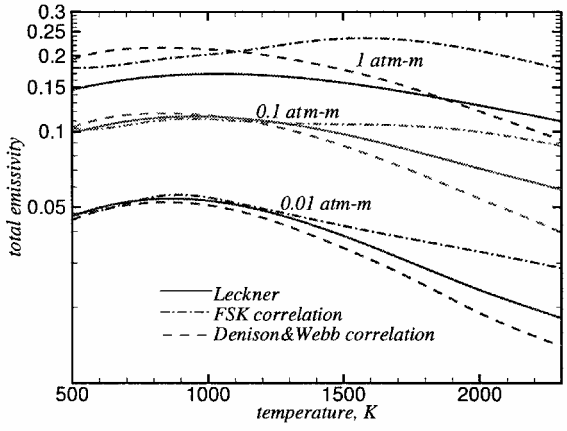

Fig. 4 Total emissivities of CO_2 from Leckner,[18] FSSK correlation, and Denison and Webb's correlation.

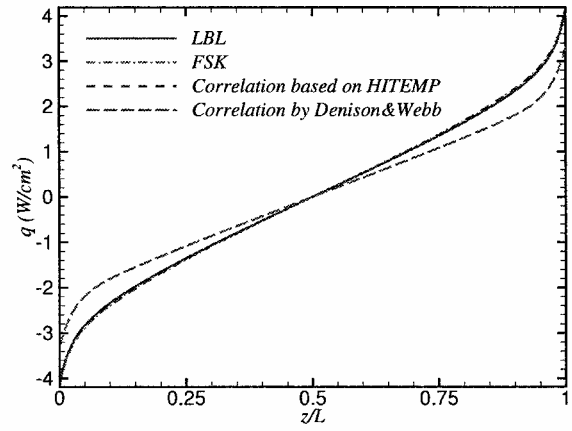

Fig. 5 Local radiative flux in an isothermal N_2–CO_2 mixture (T = 1500 K, p = 1 bar, x_{CO_2} = 0.1, and L = 1 m) bounded by cold, black walls.

based on such data. This is done in Fig. 4, where total emissivities obtained from the present correlations are compared with those obtained from Denison and Webb's correlation[7] and those calculated from Leckner's model,[18] which is based on (fairly dated) experimental data. It is observed that, as expected, the Denison and Webb correlation underpredicts high-temperature emissivities due to missing hot lines, particularly for intermediate optical thickness $p_a L$, where p_a is the partial pressure of CO_2 and L the column width. However, it is also seen that HITEMP (and the present correlation) overpredict emissivities as compared to Leckner's model.[18] As indicated by Modest and Bharadwaj,[19] HITEMP displays some questionable behavior in the band wings of CO_2 at temperatures above 1500 K, which could explain these trends and which need to be resolved.

IV. Sample Calculations

A number of relatively simple one-dimensional examples considering CO_2–N_2 mixtures confined between two infinite parallel walls are presented to test the correlations. The P-1 approximation is employed here as the RTE solver because 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). Although the FSSK method is the most accurate method, the FSCK 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 1500 K, 1 bar, with a 10% mole fraction of CO_2. Benchmark line-by-line results for the local heat flux are shown in Fig. 5, together with FSK results and those from correlations. Note that the FSK result and the HITEMP correlation essentially coincide with the LBL results, whereas 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 (Fig. 6). The pressure and CO_2 mole fraction are constant throughout at 1 bar and 10%, respectively. The hot layer is at $T = 2000$ K and has a fixed width of 50 cm, whereas the cold layer is at 300 K and is of varying width. With FSSK results and the HITEMP correlation being within 10% of the LBL result, the Denison and Webb correlation has a maximum error of 60%. Many hot lines have been activated at this high temperature of 2000 K and have not been considered in the Denison and Webb correlation.

The preceding example with a step change in temperature was designed to be a worst-case scenario. As a final 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 1-m-thick gas layer with parabolic temperature and mole fraction profiles. The black walls are kept at 300 K. In this problem, the direct FSSK shows a maximum error of about 10% and, due to compensating errors, the present approximate k-distributions

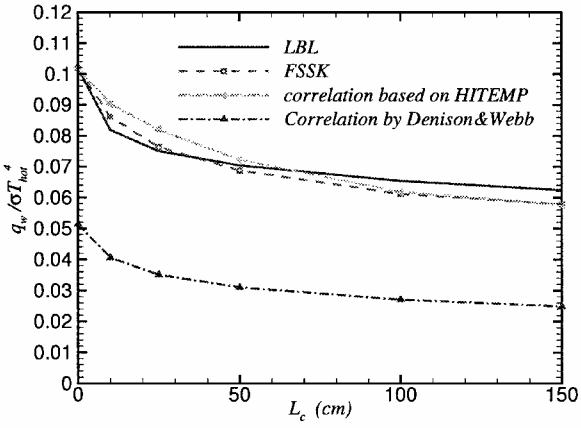

Fig. 6 Radiative flux exiting from the cold column of a two-column CO_2–N_2 mixture at different temperatures (T_h = 2000 K and L_h = 50 cm and T_c = 300 K, L_c variable, with uniform p = 1 bar, x_{CO_2} = 0.1, and cold and black walls on both sides).

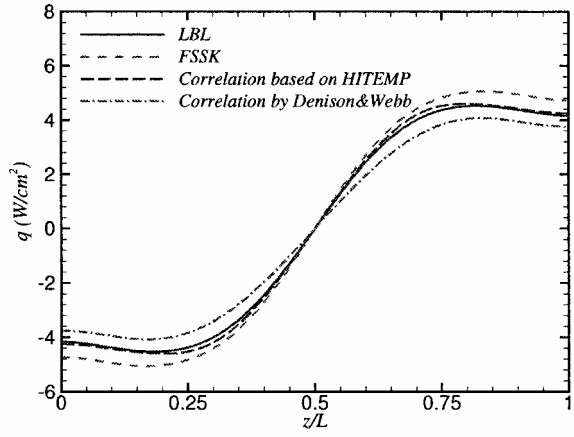

Fig. 7 Radiative heat flux in a L = 1 m thick CO_2–N_2 mixture with parabolic temperature and mole fraction profiles [$T = T_c - (T_c - T_w)(2z/L - 1)^2$, $x_{CO_2} = x_c - (x_c - x_w)(2z/L - 1)^2$, T_c = 1750 K, T_w = 300 K, x_c = 0.2, and x_w = 0.01].

reduce this error to about 5%. The Denison and Webb correlation, although also benefiting from this compensating error, overshoots the LBL results by a maximum of about 15%.

V. Summary

An approximate correlation for FSK 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 FSK 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.

Acknowledgments

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