

Narrow-Band Based Multiscale Full-Spectrum k -Distribution Method for Radiative Transfer in Inhomogeneous Gas Mixtures

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The full-spectrum k -distribution (FSK) method has become the most promising model for radiative transfer in participating media since its introduction a few years ago. It achieves line-by-line (LBL) accuracy for homogeneous media with only a tiny fraction of LBL's computational cost. Among the variants of the FSK method for dealing with inhomogeneous media, the multiscale FSK (MSFSK) method not only provides a strategy to treat the inhomogeneity problem by introducing an overlap coefficient, it also accommodates a solution to the so-called mixing problem (mixing of k -distributions for different gas species). The evaluation of MSFSK parameters, however, is tedious and excludes the MSFSK method from practical applications. In this paper a new scheme of evaluating k -distributions and overlap coefficients from a database of narrow-band k -distributions is formulated, treating each gas specie as a single scale. The new scheme makes the MSFSK method efficient and convenient for practical applications, and ready to accommodate nongray absorbing particles (such as soot) in the medium. The method virtually eliminates errors caused by uncorrelatedness due to independently varying species concentrations. It was also found that, in addition, breaking up a gas mixture into gas scales reduces the error caused by temperature inhomogeneities. The mathematical development of the new scheme is described and validated; the concept and the implication of the overlap coefficient are discussed. Sample calculations for inhomogeneous media with step changes in species mole fraction and temperature are performed to demonstrate the accuracy of the new scheme by comparison with LBL calculations. [DOI: 10.1115/1.1925281]

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1 Introduction

The concept of reordering absorption coefficient has greatly reduced the computational cost in predicting radiative transfer in gas mixtures [1,2]. Several models have been put forward to apply the concept to the entire spectrum and these include the spectral-line-based weighted-sum-of-gray-gases (SLW) model [3,4], the absorption distribution function (ADF) method [5,6], and the recent full-spectrum k -distribution (FSK) method [7]. Whereas the SLW and ADF methods are weighted-sum-of-gray-gases approaches (i.e., the—assumed to be correlated—absorption coefficient is reduced to a few discrete values) the FSK method distinguishes itself in that it is an *exact* method for a correlated absorption coefficient, utilizing a continuous k -distribution over the whole spectrum, and this allows a quadrature scheme of arbitrary order of accuracy to be employed. The FSK method achieves line-by-line (LBL) accuracy for homogeneous media with only a tiny fraction of LBL's computational cost. Since its introduction, the FSK method has undergone several major developments, including the formal mathematical development of full-spectrum correlated- k (FSCK) and scaled- k (FSSK) methods [8], the proposal of a multiscale FSK (MSFSK) method [9], the development of the multigroup FSK (MGFSK) method [10], and the assembly of full-spectrum k -distributions from narrow-band k -distributions [11].

The FSCK, FSSK, MSFSK, and MGFSK methods are designed to apply the FSK scheme to radiative transfer problems in inhomogeneous media, which remains a challenging topic for reordering models and any other global methods. The challenge is that inhomogeneities in total pressure, temperature, and component gas mole fraction (partial pressure) change the spectral distribution of the absorption coefficient, which is critical to the FSK reordering process. The effect of varying total pressure on the FSK reordering process is relatively small, as evidenced by the success of applying the correlated- k method in the field of meteorology, where strong total pressure variations occur while temperatures stay relatively uniform [1,12,13]. The effect of varying temperature can be substantial, as first recognized by Rivière et al. [14–16]: Spectral lines that are negligible at room temperatures become more and more important at elevated temperatures, giving rise to the so-called “hot lines.” The effect of varying gas concentrations can also be substantial, as first recognized by Modest and Zhang [7]: At one spatial location the absorption coefficient may be dominated by one specie, but by another (with totally different spectral lines) at a different location.

The FSCK and FSSK methods can produce accurate results for media that have large variations in total pressure but small variations in temperature and partial pressure. For media that have large temperature and partial pressure variations, sophisticated MSFSK and MGFSK methods have been developed to improve the accuracy of the FSK method. The MSFSK method deals with the inhomogeneity problem by grouping individual spectral lines comprising the absorption coefficient into M separate scales according to their temperature dependence [9]. The overlap in spectrum between different scales is treated in an approximate way so

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that a number of M independent radiative transfer equations (RTEs) (instead of M^2 required for the fictitious gas approach [5,6]) need to be solved. In contrast to the MSFSK method, which organizes spectral lines into groups, the MGFSK method places spectral positions into M separate groups according to their temperature and partial pressure dependencies [10]. This avoids the problem of overlap among different groups and, therefore, also requires only M RTE evaluations but without any further approximation. However, with the MGFSK method it appears impossible to obtain k -distributions for arbitrary gas mixtures from mixing those of individual species.

The need for mixing comes from practical considerations, since it would be impossible to precalculate and database the infinite number of possible k - g distributions that are required in a practical heat transfer calculation. In practice, the k - g distributions of individual gases are precalculated and then are mixed during the calculation to obtain arbitrary mixture k - g distributions. Although the MGFSK method can achieve great accuracy for individual gases, groups from different gases are incompatible, making its application to inhomogeneous mixtures problematic. The MSFSK method, on the other hand, can treat the absorption coefficient of an individual specie in a mixture as one of its scales. The problem of mixing FSKs of individual species in a gas mixture is thus transformed into a problem of resolving the overlap among scales.

The originally formulated MSFSK method proposed an approximate but elegant solution to the overlap problem by introducing an overlap coefficient. The calculation of this overlap coefficient, however, is cumbersome, and must be obtained from a spectral database for every location in an inhomogeneous medium. Therefore, the MSFSK method in its original form is not useful for practical calculations. It is the purpose of the present paper to develop a scheme to calculate the overlap coefficient efficiently from a database of narrow-band k -distributions, rather than from high-resolution spectral databases. This will allow the MSFSK parameters to be calculated on-the-fly, making the MSFSK method the most powerful method to deal with radiative transfer in strongly inhomogeneous gaseous media. In addition, the new scheme described here makes the MSFSK method ready to accommodate nongray absorbing particles (such as soot) in the medium. The original development of the MSFSK method does not consider nongray particles, and in addition, the boundaries were treated as black and cold. These issues will be addressed in a follow-up paper.

2 Theoretical Development

In the following, the original MSFSK method is reformulated for the convenience of further development. Many key points are clarified and improvements are indicated. All developments are in context of a radiatively participating medium contained in an opaque enclosure.

2.1 MSFSK Formulation for Gas Mixture. Although the following development can be easily extended to include gray absorbing and scattering particles as done in the original MSFSK development, for clarity, a medium consisting of a mixture of molecular gases is considered, and the radiative transfer equation (RTE) is then written as [17]

$$\frac{dI_\eta}{ds} = \kappa_\eta(\underline{\phi})(I_{b\eta} - I_\eta), \quad (1)$$

subject to the boundary condition

$$\text{at } s = 0: \quad I_\eta = I_{w\eta} \quad (2)$$

Here, I_η is the spectral radiative intensity, κ_η the absorption coefficient, $I_{b\eta}$ the spectral blackbody intensity (or Planck function), and wave number η is the spectral variable. The term $\underline{\phi}$ is a vector of state variables that affect κ_η , which include temperature T , total pressure P , and gas mole fractions x : $\underline{\phi} = (T, P, x)$. The boundary intensity $I_{w\eta}$ is due to the emission and/or reflection from the

enclosure wall. If we separate the contributions to κ_η from the M component gases and break up the radiative intensity I_η accordingly, i.e.,

$$\kappa_\eta = \sum_{m=1}^M \kappa_{m\eta}, \quad I_\eta = \sum_{m=1}^M I_{m\eta}, \quad (3)$$

then the RTE Eq. (1) is transformed into M component RTE's, one for each gas or scale. In this paper we will deal only with the treatment of concentration inhomogeneities, i.e., each gas specie will be treated as a single scale. Then the RTE for each gas scale is

$$\frac{dI_{m\eta}}{ds} = \kappa_{m\eta}(\underline{\phi})I_{b\eta} - \kappa_\eta(\underline{\phi})I_{m\eta} \quad \text{for } m = 1, \dots, M. \quad (4)$$

It is observed, physically, the intensity $I_{m\eta}$ for the m th scale is due to emission from the m th gas specie but subject to absorption from all gases.

We now apply the FSK scheme [8] to the RTE of each scale: First Eq. (4) is multiplied by Dirac's delta function $\delta(k_m - \kappa_{m\eta}(\underline{\phi}_0))$, followed by division with

$$f_m(T_0, \underline{\phi}_0, k_m) = \frac{1}{I_b(T_0)} \int_0^\infty I_{b\eta}(T_0) \delta(k_m - \kappa_{m\eta}(\underline{\phi}_0)) d\eta, \quad (5)$$

where, $\underline{\phi}_0$ and T_0 refer to a reference state. The resulting equation is then integrated over the whole spectrum, leading to

$$\frac{dI_{mg}}{ds} = k_m a_m I_b - \lambda_m I_{mg}, \quad \text{for } m = 1, \dots, M, \quad (6)$$

where

$$I_{mg} = \int_0^\infty I_{m\eta} \delta(k_m - \kappa_{m\eta}(\underline{\phi}_0)) d\eta f_m(T_0, \underline{\phi}_0, k_m), \quad (7)$$

$$g_m = \int_0^{k_m} f_m(T_0, \underline{\phi}_0, k) dk \quad (8)$$

$$a_m = \frac{f_m(T, \underline{\phi}_0, k_m)}{f_m(T_0, \underline{\phi}_0, k_m)}, \quad (9)$$

$$\lambda_m I_{mg} = k_m I_{mg} + \int_0^\infty \left(\sum_{n \neq m} \kappa_{n\eta}(\underline{\phi}) \right) I_{n\eta} \delta(k_m - \kappa_{m\eta}(\underline{\phi}_0)) d\eta f_m(T_0, \underline{\phi}_0, k_m). \quad (10)$$

Here the correlated- k (FSCK) approach has been taken, in which the correlated- k assumption deals with the inhomogeneity due to temperature change only. This implies that $k_m = k_m(T_0, \underline{\phi}, g_m)$ is evaluated from the k -distribution of the local absorption coefficient weighted by the Planck function at the reference temperature [8]. The second term in Eq. (10) is due to the overlap of the absorption coefficient of the m th scale, $\kappa_{m\eta}$ with those of all other scales, which occurs over part of the spectrum. Physically, the overlap coefficient λ_m is a reordered absorption coefficient of the m th scale taking into account the overlap with all other scales. In the MSFSK approach, the λ_m are determined approximately, based on the argument that overlap effects between scales (individual gas species in this work) are relatively small. There are many ways of approximating λ_m . Here we follow the approach used in the original MSFSK development, that is, the overlap coefficient λ_m is determined in such a way that the emitted intensity emanating from a homogeneous layer bounded by cold black walls is predicted exactly.

In Eq. (7), the reordering is performed in terms of $\kappa_{m\eta}$ and the interaction between $\kappa_{m\eta}$ and κ_η during the reordering process is

lumped into λ_m . The reordering can also be performed in terms of κ_η , which for a homogeneous layer at temperature T leads to

$$\frac{dI_{mg}^*}{ds} = \frac{k_m^* I_b}{f(T, \underline{\phi}, k)} - k_m^* I_{mg}^*, \quad \text{for } m = 1, \dots, M, \quad (11)$$

where

$$f(T, \underline{\phi}, k) = \frac{1}{I_b(T)} \int_0^\infty I_{b\eta}(T) \delta(k - \kappa_\eta(\underline{\phi})) d\eta, \quad (12)$$

$$I_{mg}^* = \int_0^\infty I_{m\eta} \delta(k - \kappa_\eta(\underline{\phi})) d\eta f(T, \underline{\phi}, k), \quad (13)$$

$$k_m^* = \frac{1}{I_b} \int_0^\infty I_{b\eta}(T) \kappa_{m\eta} \delta(k - \kappa_\eta(\underline{\phi})) d\eta. \quad (14)$$

In Eq. (11), the interaction between $\kappa_{m\eta}$ and κ_η is lumped into k_m^* . The solutions to Eqs. (7) and (11) for a homogeneous layer at temperature T bounded by cold black walls can be obtained analytically, and the total exiting intensities at $s=L$ are

$$I_m = \int_0^1 I_{mg} dg = \int_0^\infty \frac{k_m}{\lambda_m} I_b [1 - \exp(-\lambda_m s)] f_m(T, \underline{\phi}, k_m) dk_m \quad (15)$$

and

$$I_m^* = \int_0^1 I_{mg}^* dg = \int_0^\infty \frac{k_m^*}{k} I_b [1 - \exp(-ks)] dk, \quad (16)$$

respectively. The spectrally integrated intensity, I_m , should be equal to I_m^* , and this requirement leads to

$$\lambda_m = k \quad \text{and} \quad k_m f_m(T, \underline{\phi}, k_m) dk_m = k_m^*(k) dk, \quad (17)$$

or

$$k_m^*(\lambda_m) d\lambda_m = k_m f_m(T, \underline{\phi}, k_m) dk_m. \quad (18)$$

Equation (18) provides the relationship between λ_m and k_m that is required to solve Eq. (6). One convenient way of determining λ_m is using the relationship [9]

$$\int_0^{k_m} k'_m f_m(T, \underline{\phi}, k'_m) dk'_m = \int_0^{k'=\lambda_m} k_m^*(k') dk'. \quad (19)$$

In wavenumber space this may also be expressed as

$$\int_{\eta | \kappa_{m\eta} \leq k_m} \kappa_{m\eta} I_{b\eta}(T) d\eta = \int_{\eta | \kappa_\eta \leq \lambda_m} \kappa_{m\eta} I_{b\eta}(T) d\eta \quad (20)$$

Equation (19) is an implicit equation for the $\lambda_m - k_m$ relationship. In practice, the left and right hand sides of Eq. (19) are evaluated for a set of predetermined k_m and λ_m values and the results are stored in two arrays. The corresponding λ_m values for the k_m values used in the RTE evaluations are determined by interpolation from the two arrays. The so-determined λ_m will be a function of the state variables (i.e., temperature and gas species concentration) as well as k_m (or g_m).

2.2 Evaluation of Overlap Coefficient λ_m . In the original MSFSK development, the k_m^* are calculated directly from Eq. (14), using high-resolution spectral databases, making their evaluation extremely tedious and, thus, impractical for use in general problems with strongly varying concentrations. In the present work, it will be shown how the k_m^* can be determined efficiently and accurately from a database of narrow-band (NB) k -distributions of individual species (scales). The advantages of using NB k -distributions have been demonstrated by Modest and

Riazzi [11]: Assembling mixture FS k -distributions from NB k -distributions of individual gas species mixed at the narrow-band level is more accurate than mixing entire FS k -distributions of individual species. In addition, the use of NB k -distributions of individual species allows the inclusion of nongray absorbing particles in the participating medium, and can accommodate nongray scattering and nongray walls.

Substituting Eq. (14), the right-hand side (RHS) of Eq. (19) may be rewritten in terms of narrow-band k_m^*

$$\begin{aligned} \text{RHS} &= \int_0^{k=\lambda_m} \sum_{i=1}^{N_{\text{nb}}} \frac{I_{bi}}{I_b} \frac{1}{\Delta\eta} \int_{\Delta\eta} \kappa_{m\eta} \delta(k - \kappa_\eta) d\eta dk \\ &= \sum_{i=1}^{N_{\text{nb}}} \frac{I_{bi}}{I_b} \int_0^{k=\lambda_m} k_{m,i}^*(k) dk, \end{aligned} \quad (21)$$

where $k_{m,i}^*$ is the narrow-band counterpart of k_m^* , N_{nb} is the number of narrow-bands comprising the entire spectrum, and the NB Planck function I_{bi} is defined as

$$I_{bi} = \int_{\Delta\eta} I_{b\eta} d\eta. \quad (22)$$

As always in the NB k -distribution approach, we have assumed that $I_{b\eta}$ is constant over $\Delta\eta$ and can be approximated by $I_{bi}/\Delta\eta$.

In order to evaluate the integrals involving $k_{m,i}^*$ in Eq. (21) in terms of NB k -distributions, we consider the quantity Q_m

$$Q_m = \frac{1}{\Delta\eta} \int_{\Delta\eta} \kappa_{m\eta} \exp(-\kappa_\eta L) d\eta \quad (23)$$

for the i th narrow band. Physically, Q_m is related to narrow-band emission from scale m , attenuated over path L by the entire gas mixture. Q_m can be rewritten as

$$\begin{aligned} Q_m &= \frac{1}{\Delta\eta} \int_{\Delta\eta} \kappa_{m\eta} \int_0^\infty \exp(-kL) \delta(k - \kappa_\eta) dk d\eta \\ &= \int_0^\infty k_{m,i}^* \exp(-kL) dk = \mathcal{L}(k_{m,i}^*), \end{aligned} \quad (24)$$

i.e., Q_m is the Laplace transform of $k_{m,i}^*$.

Modest and Riazzi [11] have shown that, on a narrow-band basis, the spectral behavior of different species is essentially statistically uncorrelated. With this assumption, Q_m can also be written as

$$\begin{aligned} Q_m &= \frac{1}{\Delta\eta} \int_{\Delta\eta} \kappa_{m\eta} \exp(-\kappa_{m\eta} L) \prod_{n \neq m} \exp(-\kappa_{n\eta} L) d\eta \\ &\approx \frac{1}{\Delta\eta} \int_{\Delta\eta} \kappa_{m\eta} \exp(-\kappa_{m\eta} L) d\eta \prod_{n \neq m} \left(\frac{1}{\Delta\eta} \int_{\Delta\eta} \exp(-\kappa_{n\eta} L) d\eta \right). \end{aligned} \quad (25)$$

The second step follows by recognizing that the integration in the first step is an averaging operator, together with the assumption that the absorption coefficients of different scales are statistically uncorrelated. The k -distribution method can then be applied to Eq. (25) and we obtain

$$\begin{aligned} Q_m &\approx \int_0^1 k_{m,i} \exp(-k_{m,i} L) dg_m \prod_{n \neq m} \left(\int_0^1 \exp(-k_{n,i} L) dg_n \right) \\ &= \int_{g_1=0}^1 \cdots \int_{g_M=0}^1 k_{m,i} \exp\left(-\sum_{n=1}^M k_{n,i} L\right) dg_1 \cdots dg_M. \end{aligned} \quad (26)$$

Equating Eqs. (24) and (26), we have

$$\mathcal{L}(k_{m,i}^*) \approx \int_{g_1=0}^1 \cdots \int_{g_M=0}^1 k_m \exp\left(-\sum_{n=1}^M k_n L\right) dg_1 \cdots dg_M, \quad (27)$$

and, using the integral property of the Laplace transform,

$$\begin{aligned} \mathcal{L}\left(\int_0^{k=\lambda_m} k_{m,i}^*(k) dk\right) \\ \approx \int_{g_1=0}^1 \cdots \int_{g_M=0}^1 k_m \frac{\exp\left(-\sum_{n=1}^M k_n L\right)}{L} dg_1 \cdots dg_M. \end{aligned} \quad (28)$$

Finally, taking the inverse Laplace transform, we obtain

$$\int_0^{k=\lambda_m} k_{m,i}^*(k) dk \approx \int_{g_1=0}^1 \cdots \int_{g_M=0}^1 k_m H\left(k - \sum_{n=1}^M k_n\right) dg_1 \cdots dg_M, \quad (29)$$

where H is the Heaviside step function.

The LHS of Eq. (19) is also readily expressed in terms of NB k -distributions,

$$\begin{aligned} \text{LHS} &= \int_0^{k_m} k_m \frac{1}{I_b} \int_0^\infty I_{b\eta} \delta(k_m - \kappa_{m\eta}) d\eta dk_m \\ &= \sum_{i=1}^{N_{nb}} \frac{I_{bi}}{I_b} \int_0^{k_m} k_m \frac{1}{\Delta\eta} \int_{\Delta\eta} \delta(k_m - \kappa_{m\eta}) d\eta dk_m \\ &= \sum_{i=1}^{N_{nb}} \frac{I_{bi}}{I_b} \int_0^{g_{m,i}(k_m)} k_{m,i} dg_{m,i}. \end{aligned} \quad (30)$$

Equating the LHS and RHS, we obtain a generic expression for the determination of the overlap coefficient λ_m based on NB k -distributions of individual gases as

$$\begin{aligned} \sum_{i=1}^{N_{nb}} \frac{I_{bi}}{I_b} \int_0^{g_{m,i}(k_m)} k_{m,i} dg_{m,i} \\ = \sum_{i=1}^{N_{nb}} \frac{I_{bi}}{I_b} \int_{g_{1,i}=0}^1 \cdots \int_{g_{M,i}=0}^1 k_{m,i} H \\ \times \left(\lambda_m - \sum_{n=1}^M k_{n,i}\right) dg_{1,i} \cdots dg_{M,i}, \quad \text{for } m = 1, \dots, M. \end{aligned} \quad (31)$$

For special cases, for example, for a mixture of two gases, the $\lambda_1 - k_1$ relation for gas 1 can be found from

$$\sum_{i=1}^{N_{nb}} \frac{I_{bi}}{I_b} \int_0^{g_{1,i}(k_1)} k_{1,i} dg_{1,i} = \sum_{i=1}^{N_{nb}} \frac{I_{bi}}{I_b} \int_{g_{1,i}=0}^1 k_{1,i} g_{2,i} (\lambda_1 - k_{1,i}) dg_{1,i}, \quad (32)$$

and similarly for gas 2.

In the narrow-band k - g database published by Wang and Modest [18], the data are organized in such a way that the narrow-band k - g pairs obtained from the database for a certain condition correspond to the quadrature points of a Gaussian quadrature scheme. Therefore, the integrals in Eqs. (31) and (32) can be evaluated efficiently based on the narrow-band database.

2.3 Uncorrelatedness of Gas Absorption Coefficients.

In order to reach the result of Eq. (31), the absorption coefficients of different scales (component gases) have been assumed to be uncorrelated on a narrow-band basis, Eq. (25). Taine and Soufiani [19] have discovered that multiplying the transmissivities of two

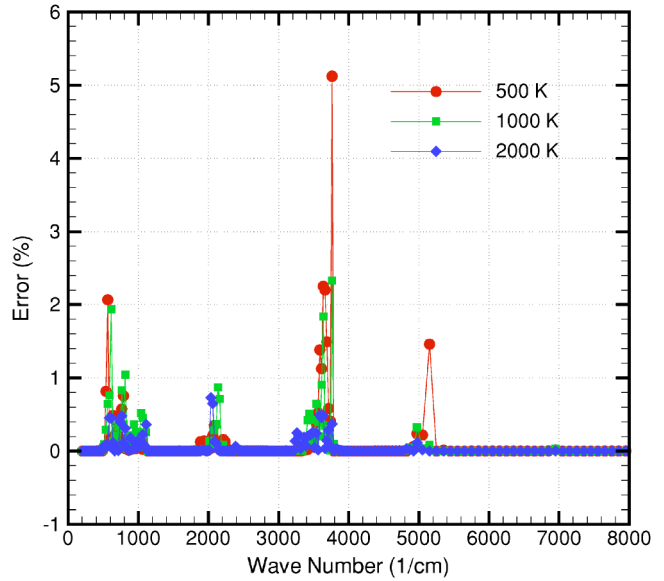


Fig. 1 Error introduced by Eq. (33) for a mixture of 10% CO₂-10% H₂O-80% N₂ at 1 bar

individual gas species to obtain a binary mixture transmissivity is very accurate for nearly all spectral locations, i.e.,

$$\begin{aligned} \frac{1}{\Delta\eta} \int_{\Delta\eta} \exp[-(\kappa_{1\eta} + \kappa_{2\eta})L] d\eta \\ \approx \frac{1}{\Delta\eta} \int_{\Delta\eta} \exp(-\kappa_{1\eta}L) d\eta \frac{1}{\Delta\eta} \int_{\Delta\eta} \exp(-\kappa_{2\eta}L) d\eta. \end{aligned} \quad (33)$$

Equation (33) does not imply that the absorption coefficients of the two gases are statistically uncorrelated, but if they are uncorrelated, Eq. (33) will follow. Figure 1 shows the relative errors of using the product of individual gas transmissivities as the mixture transmissivity when compared to the directly calculated mixture transmissivity from the LBL database. In the figure, the mixture contains 10% H₂O and 10% CO₂, and the HITEMP [20] and CDSD [21] spectral databases are used to evaluate the absorption coefficients of the two gas species, respectively. Following Wang and Modest [18], a total number of 248 narrow bands are considered, and the spectral range of each narrow band is selected such that the Planck function is nearly constant over the range (within 0.5% error for temperatures from 300 to 2500 K). The path length L is determined in such a way that the mixture transmissivity is around 0.5 for each narrow band, except for those cases where the length would be too large or too small (allowing lengths between 0.01 and 1000 cm). Each symbol in the figure represents the error for one narrow band. The errors stay below 1% for most NBs and this is true for all mixture temperatures. The error becomes smaller as the mixture temperature is increased, suggesting that Eq. (33) is more accurate for high temperature situations.

In order to ascertain the accuracy of Eq. (31), the uncorrelatedness between gas species will be tested by considering a binary mixture of the most important combustion gases, H₂O and CO₂. Equation (25) for a two-gas mixture reduces to

$$\begin{aligned} \frac{1}{\Delta\eta} \int_{\Delta\eta} \kappa_{1\eta} \exp[-(\kappa_{1\eta} + \kappa_{2\eta})L] d\eta \\ \approx \frac{1}{\Delta\eta} \int_{\Delta\eta} \kappa_{1\eta} \exp(-\kappa_{1\eta}L) d\eta \frac{1}{\Delta\eta} \\ \times \int_{\Delta\eta} \exp(-\kappa_{2\eta}L) d\eta. \end{aligned} \quad (34)$$

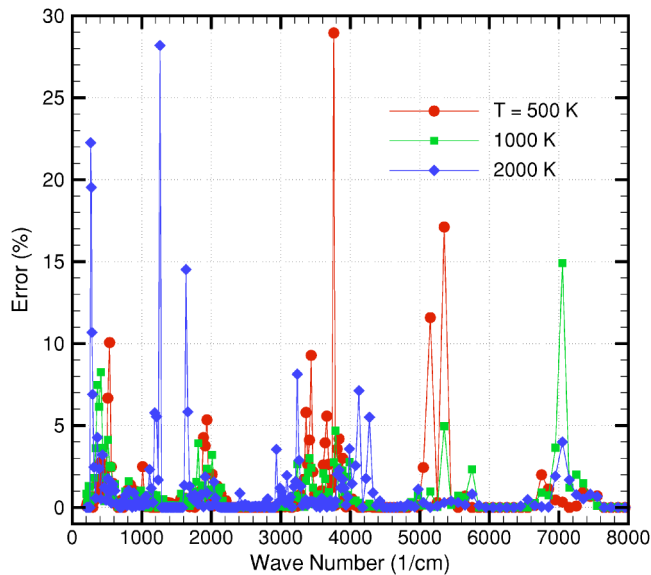


Fig. 2 Error introduced by Eq. (34) for a mixture of 10% CO₂-10% H₂O-80% N₂ at 1 bar

This relation differs from Eq. (33) in that the absorption coefficient of one gas appears in front of the exponential in the integrand of the LHS; the first part of the RHS can be recognized as the derivative of the transmissivity of gas 1 with respect to the path length. Equation (34) is exact if $\kappa_{1,\eta}$ and $\kappa_{2,\eta}$ are statically uncorrelated. The errors introduced by this approximation for all the narrow bands are shown in Fig. 2, using the same parameters as those for Fig. 1. The errors are considerably larger than the errors shown in Fig. 1, indicating that the absorption coefficients of different gases are not truly uncorrelated. The maximum errors for different mixture temperatures occur at different spectral locations. Since the introduced error will be weighted by the Planck function [RHS in Eq. (31)], the overall error from using Eq. (31) to determine the overlap coefficient λ_m is still expected to be relatively small. This is shown to be true in the following validating calculations.

2.4 Validation of Approach. To test the new scheme of evaluating the overlap coefficient from narrow-band k -distributions for individual species, the radiative intensity emerging from a homogeneous gas layer bounded by cold black walls is calculated in three ways: Using LBL calculations, using the MSFSK method with Eqs. (14) and (19) to determine the overlap coefficient directly from the HITEMP and CDSD databases (the original MSFSK method), and, finally, using the MSFSK method, but employing Eq. (31) to determine the overlap coefficient (present approach), with the required narrow-band k -distributions obtained also from HITEMP and CDSD. In the MSFSK calculations, all (5000) k -g points have been used for numerical integration, and, therefore, the results from the original MSFSK are as “exact” as the LBL calculations (which were obtained using trapezoidal rule quadrature with 1 million data points) for such a homogeneous layer and the differences between the two come only from numerical inaccuracies. The present approach contains the approximation made in Eq. (25) and differences from the LBL calculations show the error introduced by this simplification and, consequently, the validity of the new scheme to determine the overlap parameter.

Figure 3 shows the relative errors of the two MSFSK calculations compared to LBL, with errors plotted against the path length of the homogeneous layer for several temperatures. For the original MSFSK method, in which the overlap coefficient is determined “exactly,” the error increases with path length, but at a slow

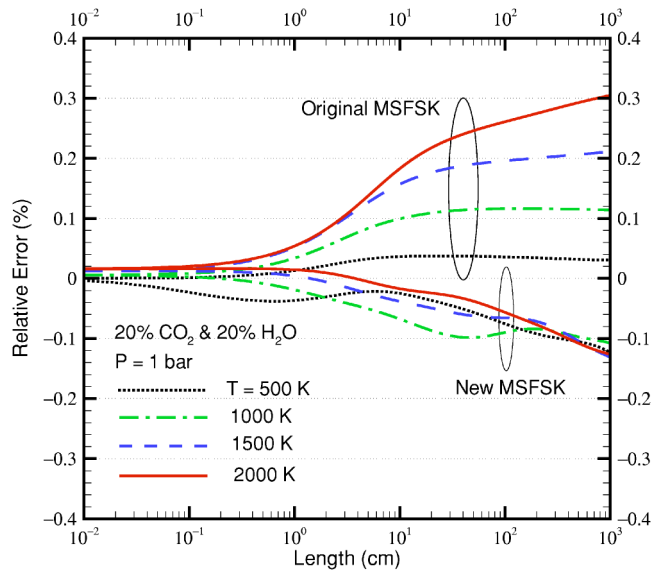


Fig. 3 Validation of the new MSFSK scheme

rate; the error also increases with temperature. Since the method is exact, the error comes only from numerical inaccuracies in the evaluation of k -distributions. For the new MSFSK implementation, in which the assumption of uncorrelated absorption coefficients between species is introduced, the error also increases with path length, but the variation with path length and temperature is not monotonic and smooth. This error comes from two sources: one is the error introduced by the uncorrelated-absorption-coefficient assumption (Fig. 2), which is weighted by the Planck function; the other is the numerical error introduced by assembling the LHS and RHS of Eq. (31) from narrow-band data. Overall, the error using the new MSFSK implementation is equivalent to the error using the original MSFSK method, proving the new scheme’s validity.

2.5 Overlap Coefficient λ_m . Figures 4 and 5 show the $\lambda_m - \lambda_m$ relations of CO₂ and of H₂O for the same homogeneous binary mixture as used in Fig. 3. As seen from Eq. (10), λ_m is the sum of k_m and the contribution from overlap with all other scales, i.e., $\lambda_m > k_m$ always. For example, Fig. 4 shows the overlap coef-

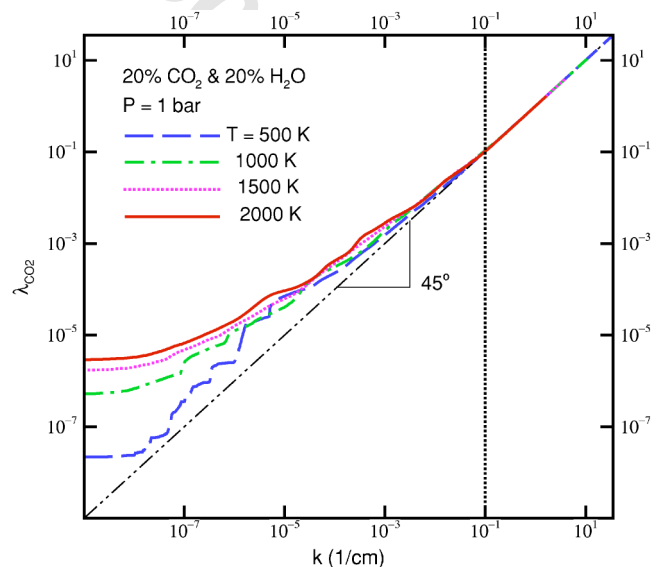


Fig. 4 $\lambda_m - k_m$ relation for CO₂

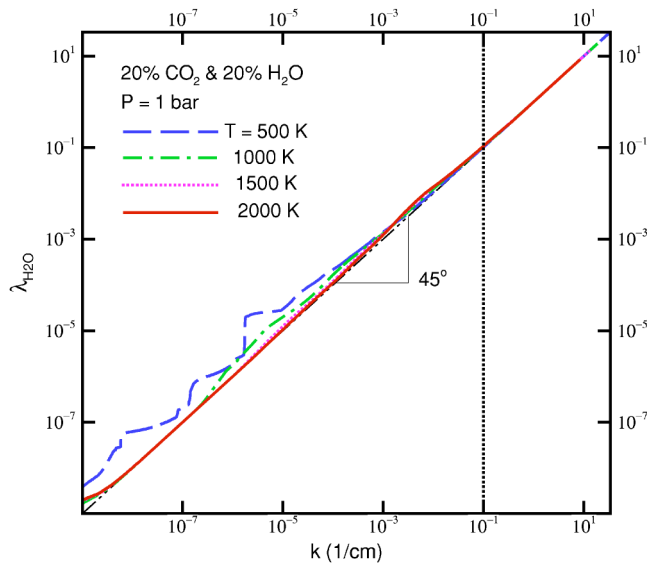


Fig. 5 $\lambda_m - k_m$ relation for H₂O

efficient λ for CO₂ as the k -distribution of CO₂ plus the overlap contribution from H₂O. Both figures show that overlap is appreciable for k -values less than about 0.1 cm⁻¹ and becomes more and more prominent as k -values decrease. This is understood by realizing that overlap occurs mostly in the wings of spectral lines. It is seen that the λ for CO₂ increases with temperature, since more and more spectral lines appear at elevated temperatures. The λ for H₂O, however, decreases with temperature. It is known that H₂O spectral lines exist almost everywhere across the entire spectrum, while CO₂ spectral lines occupy only small parts (bands) of the spectrum. At high temperatures the spectrum becomes more and more populated while the spectral lines become narrower and narrower. The portion of the H₂O spectrum that is overlapped by CO₂ apparently becomes smaller and smaller, as indicated in the figure. It is tempting to consider that for H₂O the overlap with other gases may be neglected within certain tolerance limits.

To explore the idea of neglecting overlap for H₂O, the same calculation as in Fig. 3 is performed but with overlap turned off for one or both scales (gases). The results for a mixture temperature of 1500 K are shown in Fig. 6. Even though $\lambda_{H_2O} \approx k$ (Fig. 5), not considering overlap for H₂O produces almost the same error as not considering overlap for CO₂, and is even larger at large path lengths. The explanation is found in Fig. 7, in which the k - g and λ - g distributions for H₂O and CO₂ are plotted for the same condition. The absorption coefficient of H₂O is much larger than that of CO₂ for a large part of g -space, making the small overlap in the H₂O equation more important than the overlap in CO₂.

Figure 6 also shows that the error by neglecting overlap from one or the other gas is less than about 1% for path lengths less than 10 cm, but then increases dramatically with path length. This was expected since, for path lengths less than 10 cm, the gas mixture is optically thin at nearly all wavelengths.

3 Sample Calculations

In the original development of the MSFSK method several sample calculations were conducted to demonstrate the performance of the MSFSK method. The present purpose is to improve the efficiency and convenience of the MSFSK method, by evaluating overlap coefficients and k -distributions from a narrow-band k -distribution database. In particular, we want to emphasize the accuracy of the MSFSK method in dealing with inhomogeneous media with strong partial pressure variations, i.e., in situations where the (single scale) FSK method encounters large errors.

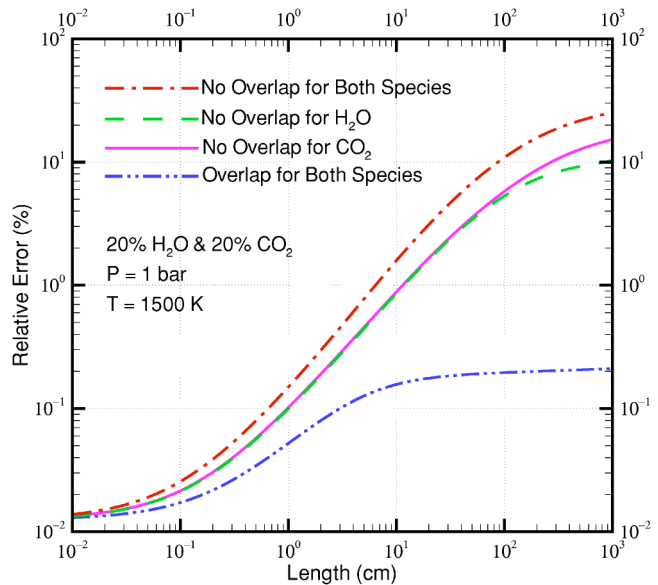


Fig. 6 Importance of neglect of overlap for one or all scales

Therefore, two one-dimensional sample calculations are considered, one with step changes in species mole fraction, the other with step changes in both species mole fraction and mixture temperature.

Both sample calculations consider a mixture of CO₂-H₂O-N₂ confined between two cold, black walls. The mixture is at a total pressure of 1 bar and consists of two different homogeneous layers (denoted as left and right layer/column). The left layer has a fixed width of $L_L = 50$ cm, and the right layer a variable width of L_R . The radiative heat flux leaving from the right layer is calculated. Five methods are used to calculate the exiting flux: The LBL, FSK (using both the correlated- k , FSCK, and the scaled- k , FSSK, approaches), and MSFSK (using correlated- k , both, with the overlap coefficient calculated directly from the spectral database, MSFSKdir, and calculated from a narrow-band database, MSFSKnb) methods. In the LBL calculations, the HITEMP and CDSO spectral databases are used for the absorption coefficients of H₂O and CO₂, respectively. In the FSK calculations, the k - g distributions are constructed directly from the spectral databases;

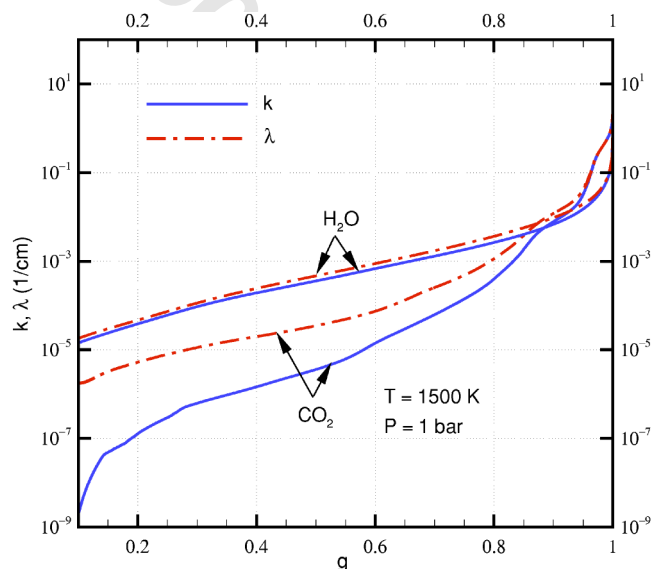


Fig. 7 k - g and λ - g distributions for water and carbon dioxide

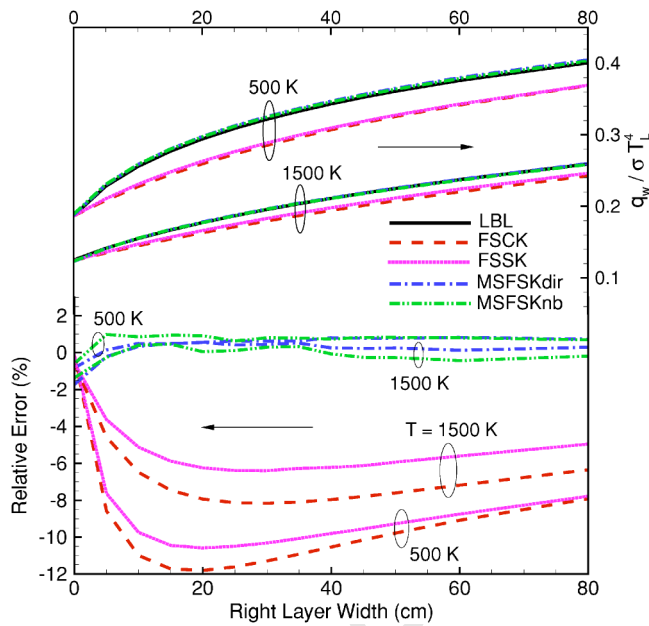


Fig. 8 Relative errors of the FSK, FSSK, MSFSKdir, and MSFSKnb calculations for step changes in mole fraction, left layer: 20% CO₂ and 2% H₂O, right layer: 2% CO₂ and 20% H₂O

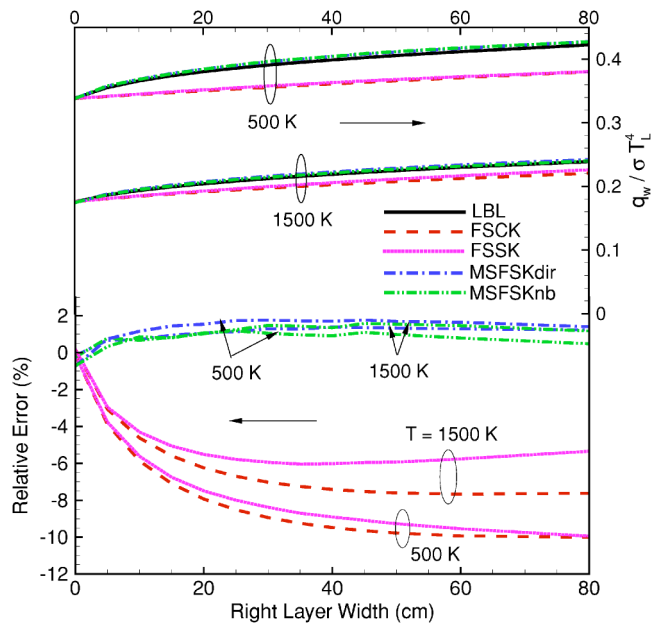


Fig. 9 Relative errors of the FSK, FSSK, MSFSKdir, and MSFSKnb calculations for step changes in mole fraction, left layer: 2% CO₂ and 20% H₂O, right layer: 20% CO₂ and 2% H₂O

the reference states and the scaling functions are determined according to the formulas in Modest and Zhang [7]. Both correlated- k and scaled- k FSK approaches are employed since, while a little more cumbersome to use, FSSK generally performs better than FSK. While both approaches can also be applied to the MSFSK methods, Zhang and Modest [22] have shown that scaled- k loses its advantages if multiple scales are used. Thus, only correlated- k is considered for the present MSFSK examples; for the MSFSKdir calculations the overlap coefficients (and k -distributions) are obtained directly from HITEMP and CDSD; for the MSFSKnb calculation the overlap coefficients and k -distributions are obtained from narrow-band k - g distributions read from the NB k - g database of Wang and Modest [18]. A ten point Gaussian quadrature scheme is used for the spectral integrations in all FSK, FSSK, MSFSKdir, and MSFSKnb calculations.

In the first example, the temperatures of both layers (T_L and T_R) are set equal, but the species mole fractions change sharply: the left layer contains 20% CO₂ and 2% H₂O, while the right layer has 2% CO₂ and 20% H₂O. The nondimensional heat flux exiting from the right layer is plotted in the upper half of Fig. 8 as a function of the right layer width (i.e., the optical thickness of the right layer). The figure includes the results for two mixture temperatures (1500 and 500 K). The heat flux increases with increasing the thickness of the right layer as the emission from the right layer builds up. The MSFSK results follow the LBL calculations closely, while the FSK methods show significant departures. The relative errors of the FSK, FSSK, MSFSKdir, and MSFSKnb calculations compared to LBL calculations are shown in the lower half of the figure. For the case of homogeneous temperature with strongly inhomogeneous gas concentration, the MSFSK method gives errors less than 1% with 10 quadrature points (and, thus, RTE evaluations) for various optical thicknesses, while the FSK and FSSK methods reach errors of 10%–12%. As the width of the right layer increases, the FSK and FSSK errors decrease, indicating that emission from the homogeneous right layer becomes dominant, and that limit can be predicted exactly by the FSK and FSSK methods. In Fig. 9, the compositions of the two layers are switched (i.e., 2% CO₂ and 20% H₂O in the left layer and 20% CO₂ and 2% H₂O in the right layer). Again, the MSFSK method gives errors less than 2% for all optical thicknesses. This sample

calculation demonstrates that in this extreme case of inhomogeneous gas concentration (partial pressure) discontinuities, the accuracy of the MSFSK method rivals that of LBL calculations, and that the new scheme of using narrow-band k -distributions for the evaluation of overlap coefficients is successful.

In the second example, in addition to a step change in species mole fraction, a step change in mixture temperature is introduced. The temperature of the left layer is set to 1500 K, and the right layer to 500 K, with the same mixture as given in Fig. 8, that is, 20% CO₂ and 2% H₂O in the left hot layer and 2% CO₂ and 20% H₂O in the right cold layer. Thus, in this example the absorption coefficients of both layers is uncorrelated due to, both, species concentration and temperature effects. Heat flux leaving the right layer is now due to emission by the left hot layer, which is attenuated by the (strongly uncorrelated) right layer. The nondimensional heat fluxes calculated by the LBL, FSK, FSSK, MSFSKdir, and MSFSKnb methods are shown in Fig. 10 against the width of the right layer. In the MSFSK calculations, the reference states are determined separately for each scale. The heat flux decreases with increasing width of the right absorption layer. For this extreme case of strong inhomogeneity in both species concentration and temperature, the FSK and FSSK methods fail completely, while the present MSFSK method, although designed for just dealing with strong inhomogeneities in gas concentration, has a maximum error of only about 12% with 10 quadrature points for various optical thickness. The difference between MSFSKdir and MSFSKnb calculations is small, indicating that the new scheme of using a database of narrow-band k -distributions to evaluate overlap coefficients is robust.

The error from the present MSFSK method can be mostly attributed to the temperature inhomogeneity and the uncorrelatedness it causes. To highlight this, the mole fraction step changes in Fig. 10 were removed and the same calculations were performed again for constant concentrations throughout. Figure 11 shows the nondimensional heat fluxes calculated by the LBL, FSK, and MSFSKnb methods for two homogeneous compositions. The differences between MSFSK and LBL calculations are similar to those in Fig. 10, in which mole fraction inhomogeneities are also present. Furthermore, it can be seen that, in addition to greatly reducing the error caused by mole fraction inhomogeneities (com-

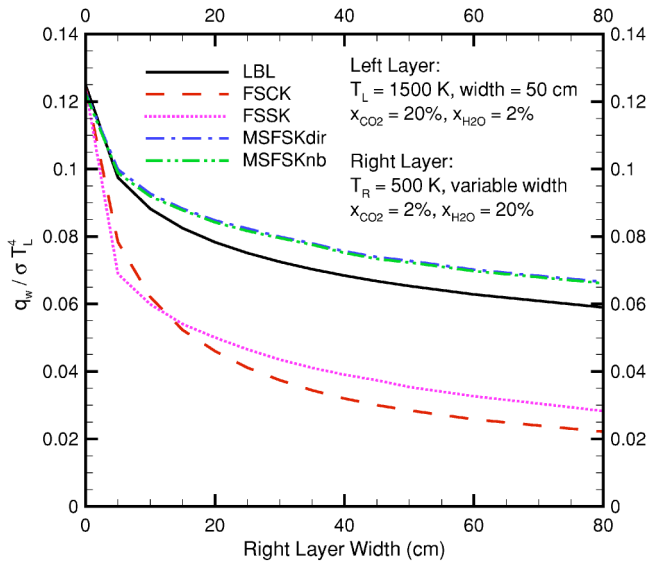


Fig. 10 Nondimensional heat flux leaving an inhomogeneous medium with step changes in species mole fraction and temperature

paring Figs. 10 and 11), the MSFSK approach (breaking up a gas mixture into gas scales) also reduces the error caused by temperature inhomogeneities: The absorption coefficient of a single gas specie is more correlated than that of a mixture that combines the spectral lines from different species. The MSFSK error can be further reduced by breaking up each gas scale into subscales to remedy temperature-caused uncorrelatedness (“hot lines”), by grouping the spectral lines of each gas according to their temperature dependencies [6,9,14]. Such breaking up into subscales in the framework of a narrow-band k -distribution database will be addressed in future work.

4 Summary and Conclusions

A new scheme has been developed and validated for the evaluation of the overlap coefficient in the multiscale full-spectrum k -distribution (MSFSK) method applied to inhomogeneous gas mixtures with each component gas treated as one scale. The new

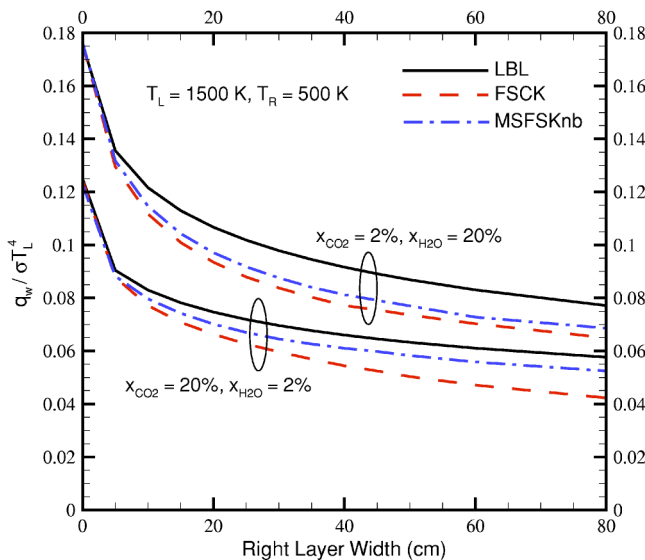


Fig. 11 Nondimensional heat flux leaving an inhomogeneous medium with step changes in temperature only

scheme exploits the uncorrelatedness of absorption coefficients of different gas species on the narrow-band level, and therefore, facilitate the use of narrow-band k -distributions. This enables the MSFSK method to be employed efficiently and conveniently in practical applications. The newly formulated MSFSK method was tested by performing radiation calculations in media with strong inhomogeneities in species mole fraction and in temperature. It was found that at isothermal conditions, the MSFSK approach essentially obtains line-by-line accuracy for inhomogeneous mixtures with extreme variations in gas concentration by treating each individual gas specie as one scale. It was also found that treating each individual gas specie as an independent scale reduces the errors caused by temperature inhomogeneities.

Acknowledgment

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Nomenclature

- a = stretching factor for FSK method
- f = k -distribution function, cm
- g = cumulative k -distribution
- H = Heaviside step function
- I = radiative intensity, W/m^2sr
- k = absorption coefficient variable, cm^{-1}
- k^* = overlap parameter defined in Eq. (14)
- L = geometric length, cm
- M = total number of scales
- P = pressure, bar
- q = radiative heat flux, W/m^2
- Q = quantity
- s = distance along path, m
- T = temperature, K
- x, \underline{x} = mole fraction (vector)
- \mathcal{L} = Laplace transform

Greek Symbols

- η = wave number, cm^{-1}
- ϕ = composition variable vector
- δ = Dirac’s delta function
- Δ = spectral interval
- λ = overlap coefficient defined in Eq. (10), cm^{-1}
- κ = absorption coefficient, cm^{-1}
- σ = Stefan–Boltzmann constant

Subscripts

- 0 = reference condition
- b = blackbody emission
- i = i th narrow band
- L = left layer
- m = m th scale
- R = right layer
- w = wall
- η = spectral in wavenumber space
- g = spectral in g space

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