APPLICATION OF THE CUMULATIVE WAVENUMBER MODEL FOR A NON-ISOTHERMAL, HOMOGENEOUS MEDIUM FILLED WITH CO₂ OR H₂O

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Abstract. This work presents the application of the cumulative wavenumber (CW) model. The CW curves are built from the HITEMP database for carbon dioxide and water vapor. The CW method replaces the highly complex dependence of the absorption coefficient with respect the wavenumber by a set of smooth curves, obtained for different absorption cross-sections. The discrete ordinates method (DOM) is used to solve the radiative transport equation (RTE) for a non-isothermal, homogeneous participating medium, and is integrated over the spectrum by means of the CW model. The behaviors of the divergent of the radiative heat flux and of the radiative heat flux are analyzed in detail. Comparisons between the HITRAN and HITEMP databases are also made in this work. The methodology is applied to a one-dimensional geometry with black walls filled with carbon dioxide or water vapor. In addition, all the results are compared with the line-by-line benchmark solution. This work shows that while the CW is a precise model to determine the divergent of the radiative heat flux, considerable errors occur for the computation of the radiative heat flux.

Keywords: Radiative Transfer, Cumulative Wavenumber, Non-isothermal problems, Spectroscopic Databases

1. INTRODUCTION

There is an increasing need for a better understanding and modeling of combustion processes, aiming at more efficient systems and at a more effective control of pollutants emission. Combustion involves a number of coupled phenomena, such as chemical kinetics, fluid flow and heat transfer. Thermal radiation in participating gases if often the dominant heat transfer mechanism in combustion due to the formation of gaseous products at high temperatures. On the other hand, the modeling is made even more difficult due to the highly complex dependence of the radiative properties with the wavelength, which can be characterized by hundreds of thousand spectral lines that represents the absorption coefficient. Thus, the solution of the radiative heat transfer equation requires special methods for the integration of the intensity field in the spectrum. An extensive overview of these models can be found in Siegel and Howell (2002).

The weighted-sum-of-gray-gases (WSGG), first proposed by Hottel and Sarofim (1967), models the entire spectrum by a few bands having uniform absorption coefficients, each band corresponding to a gray gas. The weighting coefficients that account the contribution of each gray gas correspond to the fractions of the blackbody energy in the spectrum region where the gray gases are located. In general, those coefficients are obtained from fitting experimental data, such as those presented in Smith *et al.* (1982) and Galarça *et al.* (2008), for a mixture of water vapor and carbon dioxide in nitrogen. Such data are limited, however, to a few mixtures that in addition are required to be homogeneous. The WSGG model was first proposed within the framework of the zone-method, proposed by Hottel and Cohen (1958). Modest (1991) applied the WSGG to the radiative transport equation (RTE), making it possible to use with any arbitrary solution method. Despite some important limitations of the WSGG model, this model is, together with the single gray gas model, the most largely employed method in the simulation of combustion processes, being available in most commercial codes, e. g. Fluent, CFX, StarCD, etc.

There are nowadays databases compiling characteristics related to the emission and absorption behavior of molecules, as the HITRAN, HITEMP, CDSD, among others, which have been used to obtain spectral properties of gases in much greater detail than a few decades ago. With such information, thermal radiation heat transfer can be accurately solved with the line-by-line (LBL) integration, developed by Hartmann *et al.*(1983), which considers the emission and absorption of each individual spectral line. On the other hand, LBL integrations demand a very intense computational effort, and so their use has been limited to simple geometries, especially to obtain benchmark solutions.

To avoid the difficulties related to LBL integration, various band models have been proposed. Goody and Yung (1989) proposed the Correlated-*k* assumption, which considers that, for a narrow wavenumber range the radiant energy transfer does not vary with the spectral line location, thus the lines can be rearranged for the same interval without affecting the radiative heat transfer. This assumption preserves the gas thermodynamic state, correlating the absorption coefficient in a given point of the domain to the absorption coefficient taken at the reference condition. Denison and Webb (1993a) presented the spectral-line weighted-sum-of-gray-gases (SLW) method. The modeling is based on a low

resolution spectrum using the WSGG concept. Later, Denison and Webb (1993b) applied the absorption line blackbody distribution function (ALBDF) to calculate the absorption coefficients and the weights to be used with the WSGG model. Correlations for the ALBDF were presented for H₂O and CO₂ (Denison and Webb, 1994). The SLW method leads to the appearance of Leibnitz terms in the RTE for the gray gases which makes the integration considerably more difficult. This scenario was treated by the assumption of ideal behavior of molecular spectra in temperature, assuming the correlated-k, or by simply neglecting the Leibnitz terms (Denison and Webb, 1995). The full-spectrum correlated-kdistribution (Modest, 2002) was introduced to extend the WSGG method to non-uniform media. Maurente et al. (2007) presented the application of the Monte Carlo method applied to the absorption line blackbody distribution function (MC-ALBDF) using participating media formed with water vapor and carbon dioxide. The stoichiometric ratios considered the combustion of methane and octane. Comparisons with the WSGG model were presented in Maurente et al. (2006a, 2008). Recently, the cumulative wavenumber model was proposed by Solovjov and Webb (2002) for the solution of the RTE in non-uniform gas media at high temperature. This approach allows the accommodation of nonisothermal and non-homogeneous gases, with non-gray boundaries. Solovjov and Webb (2005) applied the CW model for modeling radiative transfer in gas mixtures with soot and compared the results with LBL approach. Solovjov and Webb (2008) modeled gaseous medium using the multilayer approach by SLW and CW methods; the predictions showed high accuracy, even with few layers. Mossi et al. (2008) presented a study of the CW model application including an analysis of the molar concentration of CO₂ in a isothermal homogeneous medium. Salinas (2008) developed a fast approximate technique for the CW model. This approach reduced significantly the computational time in comparison with the standard method of solution for the CW model.

The present work presents the application of the CW modeling for a non-isothermal, homogeneous participating medium. A one-dimensional geometry with black walls filled with CO_2 or H_2O is used. A discrete ordinates method (DOM) is used to solve the radiative transport equation (RTE) taking into account the spectral dependence of the radiative properties. The radiative source and the net radiative heat flux are discussed in detail. The HITRAN and HITEMP databases are also compared. The difference of CO_2 and H_2O behavior in respect to the molar concentration is discussed. The line-by-line integration is also run for comparison.

2. MODEL FORMULATION

2.1. Spectrum Construction

The absorption cross-section of an absorbing, emitting gas is described by the Lorentz profile, which takes into account the broadening due to molecular collision. In units of $cm^2/molec.$, is given by:

$$C_{\eta} = \sum_{i} \frac{S_{i}}{\pi} \frac{\gamma_{i}}{\left(\eta - \eta_{i}\right)^{2} + \gamma_{i}^{2}}$$
(1)

where $S_i [\text{cm}^{-1}/(\text{molec.cm}^{-2}]$ is the integrated line intensity, $\eta_i [\text{cm}^{-1}]$ is the line location, and $\gamma_i [\text{cm}^{-1}/\text{atm}]$ is the half-width, which is a function of the mole fraction P_s [atm] and is described by:

$$\gamma_{i} = \left(\frac{T_{ref}}{T}\right)^{n} P_{s} \gamma_{\text{self,i}} + \left(1 - P_{s}\right) \gamma_{\text{air,i}}$$
(2)

where *T* is the temperature, γ_{self} and γ_{air} are the self broadening half-width and air broadening half-width, respectively, and *n* is the temperature dependent coefficient. The values of η_i , S_i , γ_{self} , γ_{air} and *n* can be obtained either from the HITRAN or HITEMP, among other databases.

There are some differences between these two databases: HITRAN was assembled at room temperature and the insignificant lines are neglected; HITEMP was assembled at the temperature of 1000 K, and all lines are accounted because they become important at the high temperatures that occur in most combustion processes.

The absorption coefficient of the gases, in units of cm⁻¹, is obtained by the following relation:

$$\kappa_{\eta} = \frac{7.34143 \times 10^{21}}{T} C_{\eta} \tag{3}$$

2.2. Radiative Transfer Equation

The radiative transfer equation for a non-scattering medium is given by:

$$\frac{dI_{\eta}}{dx} = -\kappa_{\eta}I_{\eta} + \kappa_{\eta}I_{b\eta} \tag{4}$$

where I_{η} is the spectral intensity at wavenumber η , $I_{b\eta}$ is the blackbody intensity, and κ_{η} is the spectral absorption coefficient. The above equation can be a priori solved for every single value of κ_{η} , but it would lead to a computational effort that would be prohibitive for most cases, with exception perhaps of one-dimensional geometries.

This work presents the solution of the radiation heat transfer in a one-dimensional slab, so that the CW model can be compared to the benchmark line-by-line solution. Thus, line-by-line predictions were obtained by dividing the spectrum into 40000 regions and a solution was carried out using the average absorption coefficient in each region. An increase to 60000 divisions into the spectrum showed no appreciable change in the results.

2.3. The CW Model

The CW model, presented by Solovjov and Webb (2002), is a non decreasing function defined by:

$$w(C,\eta) = \begin{cases} \sum_{i=1}^{L} (\beta_i - \alpha_i) + (\eta - \eta^*) \text{ if } C_\eta < C \\ \sum_{i=1}^{L} (\beta_i - \alpha_i) & \text{ if } C_\eta \ge C \end{cases}$$
(5)

where the wavenumbers α_i and β_i are obtained from the intersection of the absorption cross-sections, C_η , with the line $C_\eta = C$, and η^* is the largest wavenumber of the intersection for the case when the absorption cross-section C_η is less than the value *C*, as can be observed in Figure 1.

Differentiation of equation (5) with respect to η yields:

$$\frac{\partial w(C,\eta)}{\partial \eta} = \begin{cases} 1 \text{ for } C > C_{\eta} \\ 0 \text{ for } C \le C_{\eta} \end{cases}$$
(6)



Figure 1 - Cumulative wavenumber function (Solovjov and Webb, 2002).

Thus,

$$\int_{\{\eta:C>C_{\eta}\}} d\eta = \int_{\eta=0}^{\infty} \frac{\partial w(C,\eta)}{\partial \eta} d\eta = \int_{\eta=0}^{\infty} dw(C,\eta)$$
(7)

Next, the full spectrum, C_{η} , is subdivided into gray gases C_j , and into wavenumber subintervals H_j and D_i so that:

$$H_{j} = \left\{ \eta : C_{j-1} \le C_{\eta} \le C_{j}, \, j = 1, 2, \dots, n \right\}$$
(8)

$$\Delta_i = [\eta_{i-1}, \eta_i], \, i = 1, 2, \dots, p \tag{9}$$

One can define the set D_{ij} , named fractional gray gas, by the intersection of the sets Δ_i and H_j :

$$D_{ij} = \Delta_i \cap H_j \tag{10}$$

The consideration of local-spectrum correlation establishes that:

$$w(C_j, s, \eta) - w(C_{j-1}, s, \eta) = u_{ij}(s)v_{ij}(\eta) \text{ for } \eta \in \Delta_i$$

$$\tag{11}$$

Setting $u_{ij}(s^*) = 1$, where s^* is chosen as a reference point where the values of temperature and species concentrations are known, the function $u_{ij}(s)$ can be calculated by:

$$u_{ij}(s) = \frac{w(C_{j}, s, \eta) - w(C_{j-1}, s, \eta)}{w(C_{j}, s^{*}, \eta) - w(C_{j-1}, s^{*}, \eta)}$$
(12)

Integrating the spectral intensity of radiation I_{η} over the fractional gray gases D_{ij} , using the concept of cumulative wavenumber, yields (Solovjov and Webb, 2002):

$$\int_{D_{ij}} I_{\eta} d\eta = u_{ij}(s) \int_{\Delta i} I_{\eta} d\left[v_{ij}(\eta)\right] = u_{ij}(s) J_{ij}(s)$$
(13)

where J_{ij} is viewed as the fractional gray gas intensity, and u_{ij} as the local correction factor. The total intensity of radiation can be found by:

$$I(s) = \int_{\eta=0}^{\infty} I_{\eta}(s) d\eta = \sum_{i,j} u_{ij}(s) J_{ij}(s)$$
(14)

Considering the RTE equation in the *s* direction leads to:

$$\frac{\partial I_{\eta}}{\partial s} = -\kappa_{\eta} I_{\eta} + \kappa_{\eta} I_{b\eta} \tag{15}$$

Applying the cumulative wavenumber approach, the integration of the above RTE equation over the fractional gray gas region D_{ij} leads to:

$$\frac{\partial J_{ij}}{\partial s} = -\kappa_j J_{ij} + \kappa_j J_{bij} \tag{16}$$

The κ_j is the gray gas absorption coefficient, determined by a function of the absorption cross-section, C_j , and the molar density N (molecule/cm³), and can be determined by a geometric average between C_j and C_{j-1} :

$$\kappa_j = N_{\sqrt{C_j C_{j-1}}} \tag{17}$$

The J_{bij} is the fractional blackbody radiative energy source, and is defined by:

$$J_{bij}(s) = \int_{\Delta i} I_{b\eta} \left[T(s), \eta \right] d \left[v_{ij}(\eta) \right]$$
(18)

The summation of the $J_{bij}(s)$ gives the total emission:

$$\sum_{i,j} J_{bij} = \sigma T^4 / \pi$$
⁽¹⁹⁾

Considering $w_{C\eta}$ and $w_{YC\eta}$ the CW functions of the absorption cross-sections $C_{\eta} \in YC_{\eta}$, respectively, and neglecting the effect of self-broadening, results that the cumulative wavenumber of the spectrum YC_{η} at C is equal to the cumulative wavenumber of the spectrum C_{η} at C/Y, that is:

$$w_{YC_n}(C,\eta) = w_{C_n}(C/Y,\eta)$$
⁽²⁰⁾

3. RESULTS

3.1. Spectral Analysis

According to Solovjov and Webb (2002), the difference between HITRAN and HITEMP databases is of minor importance. Contrarily to this, studies carried out during this work revealed that there may be significant differences between the spectral lines of both databases, a discrepancy that increases with the temperature. This can be seen in Figure 1. As mentioned previously, the HITEMP data were obtained at temperature of 1000 K. Participating gases emit and absorb radiation mainly in frequencies where the corresponding photon energies match the quantum changes in energy of the gas moments, which occur more often at higher temperatures. These transitions from excited vibration levels, called "hot lines", do not appear when the spectra are obtained at the temperature of 296 K, explaining the difference in the two databases.



Figure 1: Comparison between HITRAN and HITEMP spectral databases for 100 % CO₂.

The LBL and CW used to model the absorption coefficient in the RTE consider the entire spectrum data. In combustion processes, it is recommended the use of the spectra obtained by HITEMP database due to the high temperatures that are achieved in the process. Another important point concerns the self-broadening behavior of the gases spectral lines. Solovjov and Webb (2002) presented a relation in which this term is neglected, leading to equation (20). Nevertheless, this assumption cannot be considered for all molecular gases. As seen in Fig. 2(a), the assumption described in equation (20) can be perfectly applied to the carbon dioxide, but water vapor showed a significant dependence on the self-broadening, as can be seen in Figure 2(b).



Figure 2: Dependence of the spectrum on mole fraction at 1000 K (HITEMP).

3.2. One Dimension Slab

The CW method was applied to problems involving a participating gas, either CO₂ or H₂O, mixed in an inert gas (air or nitrogen, for instance). The geometry that was considered for both cases corresponds to a one-dimensional slab, with parallel black walls placed at a distance of L=1 m. The slab is filled with a non-isothermal, homogeneous gas, according to the following cases:

Case 1: Parabolic temperature profile and medium composed of 10% CO₂.

Case 2: Cosinusoidal temperature profile and the medium composed of 10% CO₂.

Case 3: Parabolic temperature profile and medium composed of 10% H₂O.

Case 4: Cosinusoidal temperature profile and the medium composed of 10% H₂O.

The parabolic and the cosinusoidal temperature profiles are given by:

$$T(x) = 1000 - 500(2x/L-1)^{2}$$
⁽²¹⁾

$$T(x) = 1000 + 500\cos(\pi x/L)$$
(22)

The RTE solutions are performed with the discrete ordinates method, while the gas properties are modeled with the LBL and CW models. From the discrete ordinates method:

$$\mu_l \frac{dI_{\eta}^+}{dx} = -\kappa_{\eta} I_{\eta}^+ + \kappa_{\eta} I_{b\eta}$$
⁽²³⁾

$$-\mu_l \frac{dI_{\eta}^-}{dx} = -\kappa_{\eta} I_{\eta}^- + \kappa_{\eta} I_{b\eta}$$
(24)

With the LBL approach, the above equations are solved for all the lines that form the spectrum; then, the total intensity is determined by integrating the intensities over the entire spectrum. Applying the CW method, the RTE equations are applied for each fractional gray gases D_{ij} , leading to:

$$\mu_l \frac{dJ_{ij}^+}{dx} = -\kappa_j J_{ij}^+ + \kappa_j J_{bij}$$
⁽²⁵⁾

$$-\mu_{i}\frac{dJ_{ij}^{-}}{dx} = -\kappa_{j}J_{ij}^{-} + \kappa_{j}J_{bij}$$
(26)

The walls are assumed black in all cases, so the boundary conditions are:

$$J_{ij}^{+}(0,\mu_{l}) = J_{bij}(0)$$
⁽²⁷⁾

$$J_{ij}^{-}(L,\mu_l) = J_{bij}(L)$$
⁽²⁸⁾

Using the calculated fractional gray gas intensities, the net radiative flux, q''(x), and the radiative dissipation source, -dq''/dx, can be calculated, respectively, by:

$$q''(x) = \sum_{i,j} q''_{ij}(x) \text{ where } q''_{ij}(x) = 2\pi \sum_{l} \mu_{l} w_{l} \Big[J^{+}_{ij}(x,l) - J^{-}_{ij}(x,l) \Big]$$
(29)

$$-\frac{dq''}{dx} = Q(x) = \sum_{i,j} Q_{ij}(x) \text{ where } Q_{ij}(x) = 2\pi\kappa_j(x) \sum_{l} \left[w_l \left(J_{ij}^+(x,l) + J_{ij}^-(x,l) \right) \right] - 4\pi\kappa_j(x) J_{bij}(x)$$
(30)

For all cases, the CW model is compared to the benchmark solutions (LBL). The reference states chosen for the CW model are the minimum and maximum values for each temperature profile. The HITEMP database was applied since it provides more accurate spectral data when considering high temperatures, such as in the proposed cases.

First, the two cases involving a homogeneous CO_2 medium are analyzed, as described in Cases 1 and 3. The divergent of the radiative heat flux and the radiative heat flux are plotted in Figs. 3 and 4, respectively.



Figure 3: Divergent of the radiative heat flux for different reference states. The temperature profiles are given by: (a) Equation (21); and (b) Equation (22).

As shown in Figure 3(a), the CW method presents a very good agreement with the benchmark (LBL) solution when the wall temperature (T^* =500K) is assumed as the reference state. The agreement is not so satisfactory when the highest temperature in the slab (1000 K) is taken as the reference temperature. On the other hand, Solovjov and Webb (2002) reported an agreement that was scarcely influenced by the choice of the reference condition. For the cosinusoidal temperature profile, shown in Fig. 3(b), the agreement of the CW method to the line-by-line integration proved less satisfactory.

In order to observe the conservative behavior of the model, an examination of the radiative heat flux is required. These results are presented in Figure 4(a) and 4(b). For these cases a greater disagreement to the line-by-line results was found, especially for the second one, Fig.4b. A symmetric temperature profile seems to be less affected by the assumptions of the CW modeling. On the other hand, when a cosinusoidal temperature profile was considered, the model presented a behavior that was completely different from the LBL, and was also strongly dependent on the choice of the reference condition.



Figure 4: Radiative heat flux for different reference states. The temperature profiles are given by: (a) Equation (21); and (b) Equation (22).

Cases 2 and 4 are outlined considering water vapor as the participating media. Performing the CW model with equation (20), the results for the divergent heat flux in both cases are shown in Fig. 5. Differently from Cases 1 and 3, the CW model did not present a good agreement with the LBL model. This disagreement occurred because the water vapor absorption coefficient presents a strong dependence over the self-broadening, as previously presented in Figure 2, so equation (20) cannot be applied for water vapor. The heat flux results did not differed from Cases 1 and 3 as well, where the CW heat flux curves had a strong deviation in relation with LBL results.



Figure 5: Divergent of the radiative heat flux for different reference states. Not considering the self-broadening of water vapor. The temperature profiles are given by: (a) Equation (21); and (b) Equation (22).



Figure 6: Radiative heat flux for different reference states. Not considering the self-broadening of water vapor. The temperature profiles are given by: (a) Equation (21); and (b) Equation (22).



Figure 7: Divergent of radiative heat flux for different reference states. Considering the self-broadening of water vapor. The temperature profiles are given by: (a) Equation (21); and (b) Equation (22).

In order to verify the self-broadening dependence of the H₂O, Cases 2 and 4 were reran considering the spectrum construction with 10 % of water vapor, that is, setting P_s =0.1 in equation (2). Thus, equation (20) was not applied and the calculation was carried out as for a pure substance. The results are depictured in Fig. 7 and the agreements with LBL results are much better. In Fig. 7(a), the solutions obtained with the CW model were practically independent of the reference state that was used, and they only differed in the region near the center of the medium where the deviation is about 8%. In Fig. 7(b), the results are also independent of the reference state but, in this case, choosing the reference state at the minimum temperature provided good results where the maximum deviation in the medium was less than 4%.



Figure 8: Radiative heat flux for different reference states. Considering the self-broadening of water vapor. The temperature profiles are given by: (a) Equation (21); and (b) Equation (22).

Figures 8(a0 and 8(b) show the results of the heat flux along the medium. As seen, the CW model was not able to predict well the heat flux along the medium, especially when the temperature profile is not symmetric. In all cases studied, the CW model provided good results for the divergent heat flux. The model has a weak dependence with the reference state selected, unless for those cases where the absorption coefficient has a strong self-broadening dependence, like the water vapor.

The results in this paper indicate that the CW model does not match the energy balance in non-isothermal media. This can be explained by analyzing an implicit assumption that is hidden in equations (15) and (16). In fact, integrating equation (15) over D_{ij} , results that:

$$\int_{D_{ij}(s)} \frac{\partial I_n(s) d\eta}{\partial s} = \int_{D_{ij}(s)} -\kappa_{\eta} I_n(s) d\eta + \int_{D_{ij}(s)} \kappa_{\eta} I_{bn}(s) d\eta$$
(31)

Applying a finite difference scheme in the operator $\partial/\partial s$ on the left hand side of Eq. (31) and the CW definitions on the right hand side, gives:

$$\int_{D_{ij}(s)} I_n \left(s + \Delta s/2\right) d\eta - \int_{D_{ij}(s)} I_n \left(s - \Delta s/2\right) d\eta$$

$$= -\kappa_j u_{ij} \left(s\right) J_{ij} \left(s\right) + \kappa_j u_{ij} \left(s\right) J_{bij} \left(s\right)$$
(32)

The inconsistency is found on the discretized integrated terms, which is to be calculated over the fractional gray gases $D_{ij}(s)$. Note that the location of the integrated radiative intensity terms $(I_n(s + \Delta s/2) \text{ and } I_n(s - \Delta s/2))$ are different from the integration limits $D_{ij}(s)$, which is the reason for the energy balance not be verified in non-isothermal media. The current research is now focusing on fixing this aspect of the method.

4. CONCLUSIONS

This work investigated the cumulative wavenumber (CW) method, which deals with the complex spectral dependence of the absorption coefficient in non-uniform participating gases. The method was applied to a set of onedimensional problems involving either water vapor or carbon dioxide. While the medium was supposed homogeneous for each case, the temperature varied according to a parabolic (symmetric) and to a cosinusoidal (non symmetric) profiles. The results obtained with the CW model showed a satisfactory agreement with the LBL for the divergent heat flux. The solutions presented a minor dependence on the reference state that is selected for the use of the method, in which the choice of the minimum temperature as the reference state rendered the best agreements. However, the CW method presented an important inconsistency with respect to the computation of the heat flux. This results from an implicit assumption on the integration of the radiative transfer equation of the fractional gray gases that span the spectrum. The current research is attempting to fix this limitation of the method. The results also showed that the carbon dioxide presented a weak dependence on the self-broadening, the water vapor molecules have a strong dependence on it, which must be taken into account when using the CW method.

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