

THE ACCURACY OF THE RADIATIVE HEAT TRANSFER GAS MODELS IN NON-ISOTHERMAL AND NON-HOMOGENEOUS MEDIA WITH SOOT

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***Abstract.** Predictions of the amount of pollutant gases generated in the combustion of hydrocarbons, and the total heat exchanged in the process are important information demanded in the industry nowadays. Accurate combustion models are necessary to predict the production of gases like carbon dioxide, water vapor and solid particles like soot. These gases and the soot generated, combined with the high temperatures of the process, are responsible for an important amount of the radiative heat transfer exchanged in the system. If the radiative heat transfer is not accurately predicted, it can lead to poor prediction of the temperature field and, as a consequence, the formation and distribution of the gases and the amount of soot are also affected. The modeling of the absorption coefficient of the gases is very complex, due to its non-linearity behavior. On the other hand, the absorption coefficient of the soot behaves linearly with the wavenumber, allowing a simpler approach. Depending on the amount of soot generated, the radiative heat transfer from soot can be dominant, and in this case the most sophisticated and expensive gas models can be replaced by simpler ones, without losing accuracy in the prediction of the radiative heat source. In this work, a comparison between the gray gas (GG) model, the weighted sum of grays gases model (WSGG), and the cumulative wavenumber (CW) model is made for a medium composed with fixed concentrations of carbon dioxide and water vapor and different amounts of soot. The results are compared to line-by-line calculations, to verify the accuracy of the models and find the amount of soot necessary in the system to make the radiative heat transfer from soot dominant.*

***Keywords:** Radiative Heat Transfer, Spectral Gas Models, Soot radiation.*

1. INTRODUCTION

The energy necessary to keep the industry moving and to transport people to different places comes basically from the combustion of hydrocarbons. But environmental concerns on the pollution of the gases emitted during the process are raising, so new techniques have been developed to control and reduce the levels of pollution. In this sense, good modeling of the soot formed in the process is important, because it affects the temperature field and can change the amount and the concentration of the emitted gases. In the radiative heat transfer, the emissions from the soot can easily dominate the process, so the absorption and the emission from these gases do not play an important role when soot is present. In the last few years, there has been a growing effort to predict the soot formation in combustion process. The main mechanisms of soot life are the nucleation, surface growth, agglomeration, and oxidation. Moss et al. (1988) proposed a model based on a premise that the soot reaction rates could be specified in terms of the mixture fraction. After some experiments, it was verified that the acetylene is responsible for the surface growth and that the Polycyclic Aromatic Hydrocarbons (PAH) initiate the soot nucleation. The oxidation is performed mainly by the OH particles, but O₂ molecules are important in this process and have to be considered. Thus, Fairweather et al. (1992) proposed a simplified two-equation model for soot, where their model contains rate process for the nucleation, surface growth, agglomeration and oxidation. Experiments made by Sunderland et al. (1995) showed that the parameters used by Fairweather et al. (1992) are larger than the ones obtained experimentally, which could result in a serious mistake in the amount of smoke produced by the flame. Wang et al. (2005) applied two radiation models to an oxygen-enriched, propane-fueled, turbulent, non-premixed jet flame. The results showed that soot and spectrally radiating gas-phase species were distributed separately in the flame, and this segregation of radiating media strongly affects the radiant heat flux, flame structure and flame temperature. A numerical study of combustion in a liquid rocket engine was performed by Byun and Baek (2007). The simulation takes account the spray combustion at all speeds in the rocket engine and considers a non-gray finite-volume radiation model to investigate the radiation effect in turbulent combustion conditions, adding the soot formation and its effect on the radiation and flow field. Liu et al. (2004) studied the effects of radiation and the individual influence of gas and soot radiation on soot formation in counterflow C₂H₄SF diffusion flames by comparing the numerical results against available experimental data in the literature.

Soot emits a considerable amount of radiation in comparison to participating gases such as water vapor and carbon dioxide, and it changes the temperature distribution, the concentration and formations of the all species involved in the process. This justifies the importance of an accurate modeling of radiation in media composed of soot and participating gases. In this work, it will be analyzed the influence of the soot on the radiative heat transfer. The main goal is to evaluate which radiation model is more appropriate to deal with a mixture of gases and soot, considering different amounts of soot in the medium.

2. THE ABSORPTION COEFFICIENT OF THE GASES AND THE SOOT

The absorption coefficient of the gases is known by its strong variation along the wavenumber. According to Siegel and Howell (2002), for engineering applications, the absorption coefficient of the gases can be obtained using the Lorentz collision profile, given by:

$$\kappa_{\eta} = NC_{\eta} = N \sum_i \frac{S_i}{\pi} \frac{\gamma_i}{(\eta - \eta_i)^2 + \gamma_i^2} \quad (1)$$

where N is the molar density, C_{η} is the absorption cross-section, S_i is the integrated line intensity, η_i is the line location, and γ_i is the half-width, that is defined by:

$$\gamma_i = \left(\frac{T_{\text{ref}}}{T}\right)^n Y_s \gamma_{\text{self},i} + \left(\frac{T_{\text{ref}}}{T}\right)^{0.5} (1 - Y_s) \gamma_{\text{air},i} \quad (2)$$

where Y_s is the molar fraction, T is the temperature, γ_{self} is the self broadening, γ_{air} is the air broadening half-width.

The parameters used are obtained from spectral databases like HITRAN, HITEMP, GEYSA, CDSD, etc. Figure 1 shows the absorption coefficient for 10% CO₂ at 1000 K. As can be seen, it has a strong dependence on the wavenumber.

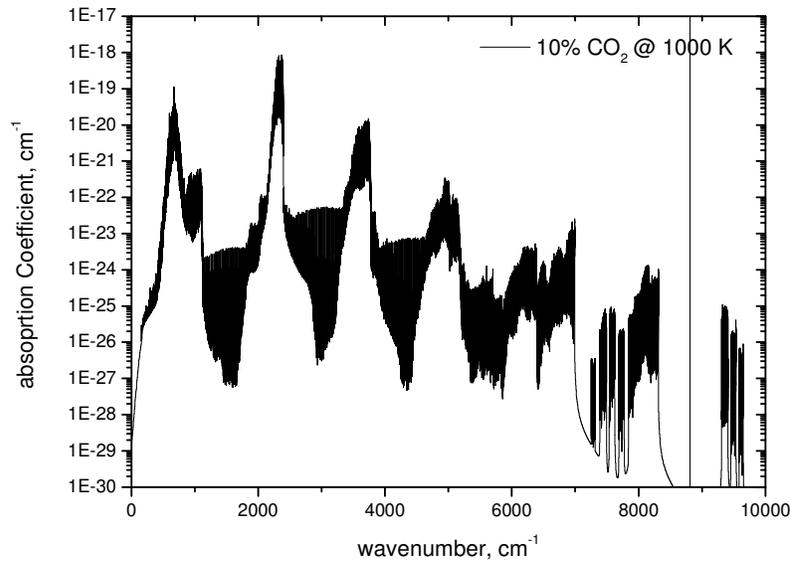


Figure 1: Absorption Coefficient of 10% CO₂ at 1000 K.

On the other hand, the absorption coefficient for the soot varies linearly with the wavenumber. According to Hottel and Sarofim (1967), it is obtained by the following relation:

$$\kappa_{\eta} = 7 f_v \eta \quad (3)$$

3. RADIATION HEAT TRANSFER MODELING

The radiative transfer equation (RTE) for non-scattering media is given by:

$$\frac{dI_\eta}{ds} = -\kappa_\eta I_\eta + \kappa_\eta I_{b\eta} \quad (4)$$

which is subjected to the boundary conditions at the walls:

$$I_{w\eta} = \varepsilon_w I_{b\eta} + \frac{(1-\varepsilon_w)}{\pi} \int_{\hat{\mathbf{n}} \cdot \hat{\mathbf{s}} < 0} I_\eta |\hat{\mathbf{n}} \cdot \hat{\mathbf{s}}| d\Omega \quad (5)$$

where ε_w is the emissivity of a diffuse gray wall and the is the spectral intensity leaving the wall due to emission and reflection.

Solving Eq. (4) for every single absorption coefficient value is a difficult task, because the absorption coefficient is strongly dependent on the wavenumber. Due to this fact, gas models have been used to solve the RTE quickly. A brief description of the models used in this work is described below.

Gray Gas Model

Many researchers, especially to solve 3D problems, have used the assumption that the gas is gray. The RTE for a non-scattering media, with the gray gas model, becomes:

$$\frac{dI}{ds} = -\kappa I + \kappa I_b \quad (6)$$

The boundary conditions can be defined as:

$$I_w = \varepsilon_w I_b(T_w) + \frac{(1-\varepsilon_w)}{\pi} \int_{\hat{\mathbf{n}} \cdot \hat{\mathbf{s}} < 0} I |\hat{\mathbf{n}} \cdot \hat{\mathbf{s}}| d\Omega \quad (7)$$

Table 1: Curve fits for the absorption coefficient used in the gray gas model

Species	Absorption Coefficients	
CO ₂ and H ₂ O	$\kappa = \sum_{i=0} c_i \left(\frac{1000}{T} \right)$ in m ⁻¹ atm ⁻¹	
		H ₂ O
	c ₀	-0.23093
	c ₁	1.1239
	c ₂	9.4153
	c ₃	-2.9988
	c ₄	0.51382
c ₅	-1.8684 × 10 ⁻³	
C	$\kappa = 1186 f_v T$ in m ⁻¹	

In this work, the absorption coefficients for CO₂, and H₂O are correlated by Barlow et al. (2001) and the absorption coefficient for the soot is calculated as suggested by Atreya et al. (1998). These curve fits are listed in Table 1.

The Weighted-Sum-of-Gray-Gases (WSGG) Model

The RTE with the WSGG model is given by:

$$\frac{dI_j}{ds} = -\kappa_j I_j + \kappa_j a_j I_b \quad (8)$$

subjected to the boundary condition:

$$I_{wj} = a_j(T_w) I_b(T_w) \quad (9)$$

where a_j is the weighting factor for the j -th gray gas. These weighting factors are defined by:

$$a_j = \int_{\Delta\eta_j} E_{b\eta}(T, \eta) d\eta / E_b(T) \quad (10)$$

The absorption coefficient κ_j is for the j -th gray gas. The total intensity can be found by summing the intensities associated with each gray gas:

$$I = \sum_{j=1}^N I_j \quad (11)$$

The WSGG model is frequently used with the correlations proposed by Smith et al. (1982) for the mixture of gases and for the soot. These correlations are listed in Table 2.

The local absorption coefficient of the j -th gray gas is obtained by the product of the modeled coefficient $\kappa_{g,j}$ by the local overall combustion products partial pressure $P_t = P_{\text{CO}_2} + P_{\text{H}_2\text{O}}$, and their weights are approximated by a cubic polynomial, given by:

$$a_{g,j} = \sum_{i=1}^4 b_{g,j} T^{i-1} \quad (12)$$

For a mixture with soot, one more gray gas with null absorption coefficient is added ($\kappa_{g,0} = 0$). Its correspondent weight is given:

$$a_{g,0} = 1 - \sum_{j=1}^3 a_{g,j} \quad (13)$$

In a case of pure gas mixtures, it does not contribute to the process, but in a mixture of gases with soot, this assumption can consider the absorbing and non-absorbing gas regions in the spectrum.

Table 2: Coefficients of the WSGG model for gas mixtures, and for the soot.

Mixture, $P_{\text{H}_2\text{O}}/P_{\text{CO}_2}=2$					
$\kappa_{g,j}$, $\text{m}^{-1}\text{atm}^{-1}$	$b_{g,j1} \times 10^1$	$b_{g,j2} \times 10^4$	$b_{g,j3} \times 10^7$	$b_{g,j4} \times 10^{11}$	
0.4201	6.508	-5.551	3.029	-5.353	
6.516	-0.2504	6.112	-3.882	6.528	
131.9	2.718	-3.118	1.221	-1.612	
Soot					
$\kappa_{s,j}$, m^{-1}	$b_{s,j1}$	$b_{s,j2} \times 10^4$	$b_{s,j3} \times 10^7$	$b_{s,j4} \times 10^{11}$	
1.00802×10^9	1.42	-7.7942	-0.38408	2.4166	
3.2352×10^9	-0.42	7.7942	0.38408	-2.4166	

For the soot, the corresponding absorption coefficient is obtained by the product of the model coefficient and the soot volume fraction, and its weighting is given in an analogous way to Eq. 11:

Thus, the absorption coefficient, used in Eq. 7 for the mixture of the gases and soot, is obtained by all possible combinations of the gas and soot absorption coefficients given in table 2, defined as:

$$\kappa_j = \kappa_{m,n} = \kappa_{g,m} + \kappa_{s,n} \quad (14)$$

and the weights are defined as:

$$a_j = a_{m,n} = a_{g,m} a_{s,n} \quad (15)$$

where the dimension of j is $m \times n$.

The Cumulative Wavenumber Model

In this model, the entire range of the absorption cross-section is divided into n gray gases, but in this model, a non-decreasing function, called the cumulative wavenumber function, is defined as:

$$w(C, \eta) = \int_0^\eta H(C - C_\eta) d\eta \quad (16)$$

where $H(C-C_\eta)$ is the Heaviside step-function, C is a gray gas, and C_η is the absorption cross-section. Differentiation of Eq. (16) with respect to η yields:

$$\frac{\partial w(C, \eta)}{\partial \eta} = \begin{cases} 1 & \text{for } C > C_\eta \\ 0 & \text{for } C < C_\eta \end{cases} \quad (17)$$

Thus, the integration of the wavenumber only in the regions where the gray gas coefficient C is bigger than the true absorption cross-section is equivalent to integrating the derivative of the cumulative wavenumber function in the entire spectrum, according to Eq. (18).

$$\int_{\eta: C > C_\eta} d\eta = \int_{\eta=0}^{\infty} \frac{\partial w(C, \eta)}{\partial \eta} d\eta = \int_{\eta=0}^{\infty} dw(C, \eta) \quad (18)$$

The cumulative wavenumber method, proposed by Solovjov and Webb (2002), can be thought as a discretization in the fractional gray gas wavenumbers (D_{ij}) space. This interval D_{ij} is defined as a intersection of two wavenumber intervals, H_j and Δ_i . The interval H_j is the wavenumber region where the absorption cross-section is between two adjacent gray gases, that is:

$$H_j = \{\eta : C_{j-1} \leq C_\eta < C_j, \quad j = 1, \dots, n\} \quad (19)$$

and the interval Δ_i is the wavenumber region divided in subintervals:

$$\Delta_i = [\eta_{i-1}, \eta_i], \quad i = 1, \dots, K \quad (20)$$

For all $\eta \in \Delta_i$, the difference between two adjacent gray gases can be viewed as a product of two functions, according to Eq. (21).

$$w(C_j, s, \eta) - w(C_{j-1}, s, \eta) = u_{ij}(s) v_{ij}(\eta) \quad (21)$$

where the function $v_{ij}(\eta)$ is the difference in the wavenumber function evaluated at a reference thermodynamic state s^* . Thus, the function $u_{ij}(s)$ can be defined as:

$$u_{ij}(s) = \frac{\Delta w(C_j, s, \eta)}{v_{ij}(\eta)} = \frac{w(C_j, s, \eta) - w(C_{j-1}, s, \eta)}{w(C_j, s^*, \eta) - w(C_{j-1}, s^*, \eta)} \quad (22)$$

Thus, the integration of the radiative spectral intensity I_η , over the fractional gray gases D_{ij} intervals, using the cumulative wavenumber approach yields:

$$\int_{D_{ij}} I_\eta d\eta = u_{ij}(s) \int_{\Delta_i} I_\eta d[v_{ij}(\eta)] = u_{ij}(s) J_{ij}(s) \quad (23)$$

where J_{ij} is viewed as a fractional gray gas intensity, and u_{ij} as a local correction to the fractional gray gas intensity. With this approach, Eq. (4) can be written as:

$$\frac{\partial J_{ij}}{\partial s} = -\kappa_j J_{ij} + \kappa_j J_{bij} \quad (24)$$

κ_j is the gray gas absorption coefficient, defined as:

$$\kappa_j = N \sqrt{C_j C_{j-1}} \quad (25)$$

and J_{bij} is the fractional blackbody radiative energy source

$$J_{bij}(s) = \int_{\Delta_i} I_{b\eta} [T(s), \eta] d[v_{ij}(\eta)] \quad (26)$$

The total intensity is obtained by the summation of the product between u_{ij} and J_{ij} over all fractional grays gases, that is:

$$I(s) = \sum_{i,j} u_{ij}(s) J_{ij}(s) \quad (27)$$

In the CW method, the spectrum is assumed to vary linearly with the concentration:

$$w_{Y_{C_\eta}}(C, \eta) = w_{C_\eta}(C/Y, \eta) \quad (28)$$

It is also assumed that the intersections of the gray gases C with the spectrum $C_\eta + C^*$ (the sum of the absorption cross-section with the absorption coefficient of gray particles), produce the same wavenumber intervals as the intersections of the gray gas $C - C^*$ with the spectrum C_η , that is:

$$w_{C_\eta + C^*}(C, \eta) = w_{C_\eta}(C - C^*, \eta) \quad (29)$$

Then, for a mixture of gases and soot, Solovjov and Webb (2002) defined some approaches that can facilitate the use of this method: the superposition approach, the multiplication approach, and the hybrid approach. In this work, is used the superposition approach, which assumes that the absorption cross-sections of the species do not overlap and that the non-gray particles are piece-wise constant in the interval Δ_i . Thus,

$$w(C, \eta) = (1-m)\eta + \sum_{k=1}^m w_k(C/Y_k - C_i^s, \eta) \quad (30)$$

where $C_i^s = C_\eta^s$ for $\eta \in \Delta_i$, and C_η^s is given by Eq. (3)

4. RESULTS

In this section, it is analyzed the influence of the soot in the radiative heat transfer. It is considered a geometry with parallel black walls placed at a distance of $L=1$ m. The slab is filled with a mixture of gases composed by 10% CO₂ and 20% H₂O, where the temperature changes according to the following equation:

$$T(x) = 1000 - 500(2x/L - 1)^2 \quad (31)$$

Four levels of soot in the medium are considered: $f_v = 1 \times 10^{-8}$, $f_v = 1 \times 10^{-7}$, $f_v = 1 \times 10^{-6}$ and $f_v = 1 \times 10^{-5}$. Figure 2 shows line-by-line calculations for the divergence of the radiative heat flux, or the volumetric radiative heat source, in this situation. As can be seen, there is no significant difference between a situation where there is no soot and a situation with a very low concentration of soot ($f_v = 1 \times 10^{-8}$), but when soot is increased to $f_v = 1 \times 10^{-7}$, an important raise occurs in the higher temperature region of the medium, but only a small variation in the colder region.

For a situation where the soot concentration is $f_v = 1 \times 10^{-6}$, it dominates the radiative heat transfer. Thus, in this case, the gas has no important contribution. And going into a non-usual situation where the soot concentration is as high as 1×10^{-5} , the medium becomes so optically thick that the divergence of the radiative heat transfer decreases. In this situation, the medium becomes almost opaque.

Simulating the same situation again, but using the gray gas model to solve the radiative heat transfer equation, it is observed no variation from a situation where there is no soot to a case where the soot concentration is $f_v = 1 \times 10^{-6}$. It shows that, when the gray gas model is used with the Planck mean absorption coefficient values, the divergence of the radiative heat flux is overestimated both in the high and low temperature regions. The difference was only observed in an extremely thick situation, where the concentration of soot in the medium is 1×10^{-5} .

Taking advantage of the fact that the absorption coefficient of the soot is easy modeled, even a simple model as the gray gas can produce good results in a situation where the radiative heat transfer from soot is dominant. But in situations where the radiative heat transfer from the gas is important, the gray gas model can lead to large errors in the calculations. Figures 4 and 5 show a comparison between the gray gas model and the line-by-line calculations, for those situations where the soot dominates the heat transfer. When the soot concentration is 1×10^{-6} , the largest difference is in the cold region, where it reaches about 30%. But in the hot region, the difference is just 3.5%.

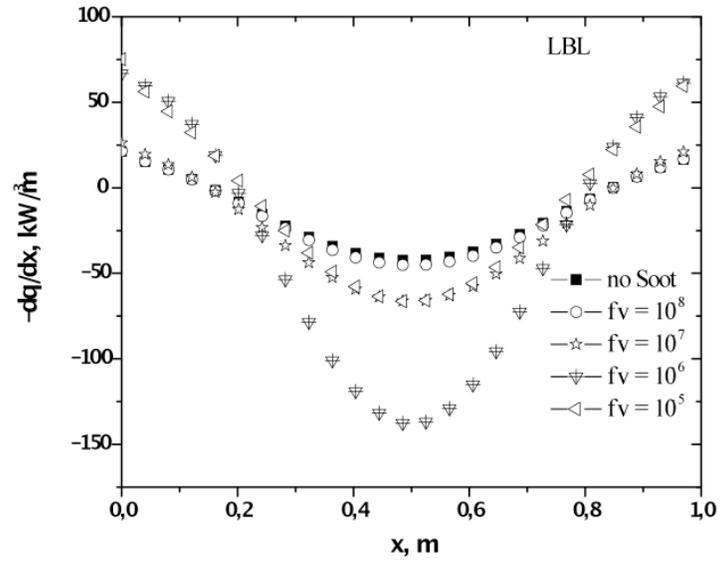


Figure 2: Benchmark results obtained with line-by-line calculations.

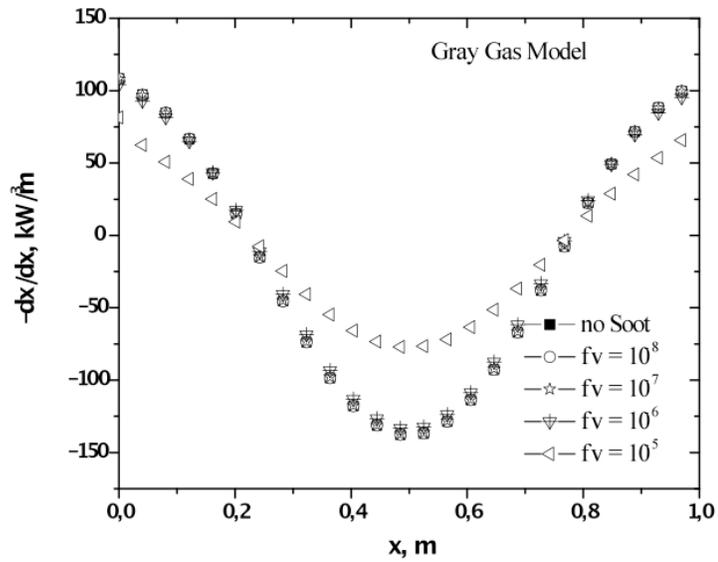


Figure 3: Results obtained with the gray gas model.

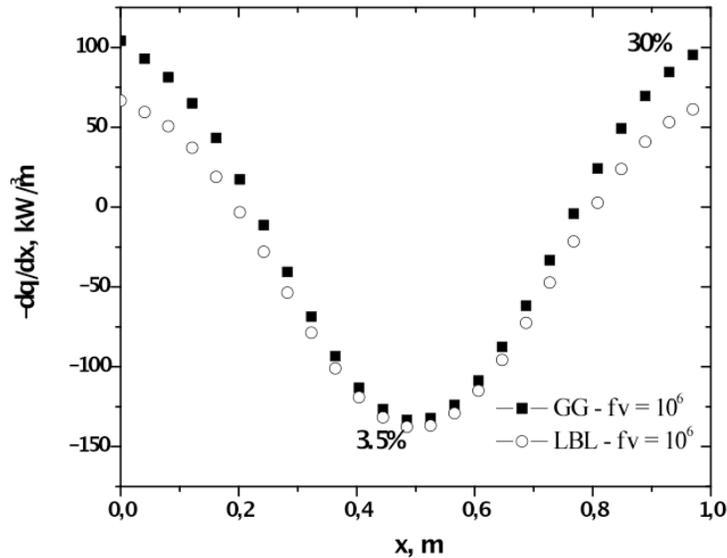


Figure 4: Comparison between the gray gas model and line-by-line calculations for a medium with 1 ppm of soot.

This difference is further reduced if the soot concentration is higher ($f_v = 1.10^{-5}$). As can be seen in Figure 5, in the cold region, the error is just 7%, and in the hot region it increases to 16%.

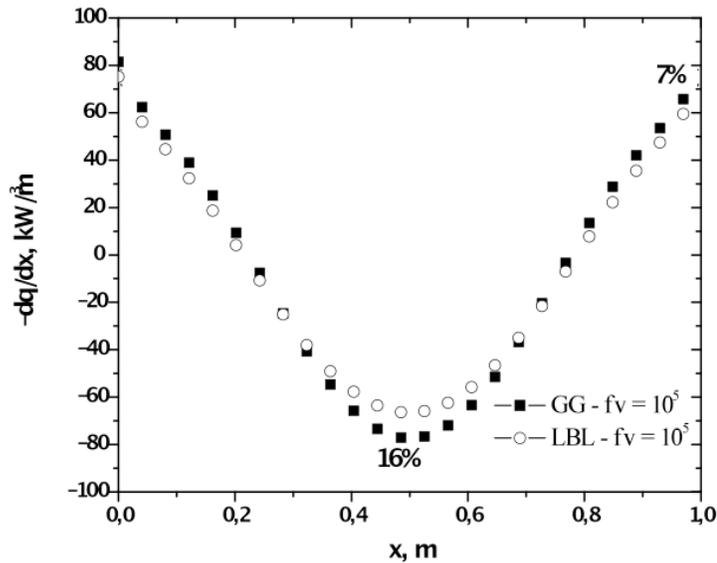


Figure 5: Comparison between the gray gas model and line-by-line calculations for a medium with 10 ppm of soot.

Other models, such as WSGG and CW, produces better results for problems where the radiative heat transfer from the gas is important. Figure 6 shows the results obtained with the WSGG model, which is described in Section 3. As can be verified, the model followed the same trend of the LBL calculations did, but the values are smaller than the one previously obtained. This behavior probably happened because the correlations used in this paper are out-of-dated, since they were obtained in the 80's. At that time, the spectral database used to obtain the absorption coefficient was in the first stage, so the values are very different from the ones used nowadays. Despite this problem, the WSGG model seems to be efficient to predict the radiative heat transfer from the gas, and it is relatively fast.

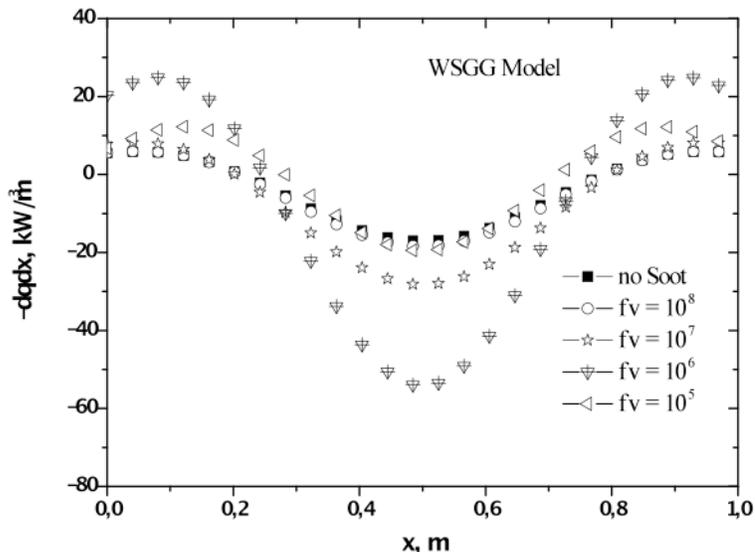


Figure 6: Results obtained with the WSGG model (the correlations proposed by Smith et al. (1982) were used)

Figure 7 depicts the results obtained with the CW model. This spectral model produces good results for the situations where the radiative heat transfer from the gas is important, but running with soot, the results do not follow the same trends. The approach used for treating the soot with this model was not efficient, especially for cases where the soot concentration is high ($f_v = 1 \times 10^{-6}$ and $f_v = 1 \times 10^{-5}$). For the case where the medium becomes extremely thick ($f_v = 1 \times 10^{-5}$), the CW model did follow the same trend as the other models did. As such, the divergence of the radiative heat flux is higher.

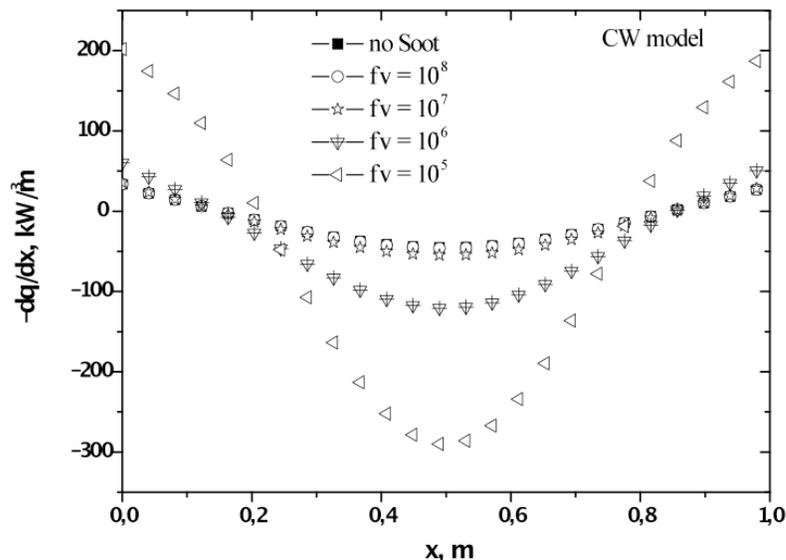


Figure 7: Results obtained with the CW model.

5. CONCLUSIONS

This work shows that in a medium composed with gases, like CO_2 and H_2O , and solid particles like soot, the models used to solve the radiative heat transfer equation must be carefully employed. For situations where the radiative heat transfer from the gases is significant, models that consider the variation of the absorption coefficient with the wavenumber produce better results. On the other hand, the correlations used with the WSGG model must be up-dated to get better results. When the levels of soot in the medium increase, the divergence of the radiative heat flux increase too, and it was verified that the radiative heat transfer from soot becomes dominant, in this case, from 1 ppm of soot. For these amounts, the gray gas model can be applied, because it produces fast and good approximations. But for situation where the radiation from the gas is important, the gray gas model does not estimate the divergence accurately.

In cases evolving combustion of hydrocarbons, soot and gases are always generated. Then the radiative heat transfer is important in the process. According to this study, the CW model is a good model to be used in the radiative heat transfer equation if the gas is dominant. For cases where the soot is dominant, the gray gas model is good because it is fast and describes the process fairly. For intermediate situations, where both the gas and the soot are important, the WSGG model is indicated, but the correlations used to make it faster must be updated.

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