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Effects of fuel absorption on radiative heat transfer in methanol pool fires

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Abstract

Spectrally resolved infrared absorption coefficients of methanol were measured using high temperature Fourier transform infrared (FTIR) spectroscopy for a range of temperatures (up to 1000 K) expected to be within the fuel rich core of pool fires. Principal absorption band peaks decreased with increasing temperature. The spectral region of the dominant C–O stretching peak causes the integrated Planck mean absorption coefficient to decrease above 350 K. Unidirectional radiation intensity along the fire centerline for a 0.3 m diameter methanol pool was calculated using the 1-D radiative transport equation with the highly resolved absorption coefficient database of methanol and combustion products, and gas-phase temperature and species profiles calculated by NIST's Fire Dynamics Simulator. The spectrally integrated intensity at the methanol pool surface was calculated at 12,400 W/m² sr, which agreed to within 2% of previously reported unidirectional radiation intensity measurements. Intensity calculations with oft-used simplifications for modeling fuel vapor absorption were compared with the spectrally resolved, temperature dependent absorption calculations, and errors with the simplified approaches for integrated radiation intensity to the pool surface were as high as 20%. This study suggests that even for small methanol pool fires, gas-phase fuel absorption coefficients should have accurate assessments for calculating radiation heat transfer to the pool surface.

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1. Introduction

Many studies have shown the importance of radiative heat transfer to the liquid surface in determining mass burning rates of pools for many scenarios including compartment fires [1], tunnel fires [2], and oil tank fires [3,4]. For laboratory scale pool fires with pool diameters, d_{pool} as small as 30 cm, radiation has been shown to be more

than half of the total heat transfer to pool surface [5] for both sooting (heptane and toluene) and non-sooting (methanol) fuels with the fraction of heat transferred via radiation increasing with sooting tendency. Furthermore, radiative heat transferred to the liquid fuel increases more rapidly with d_{pool} than convective heat transfer, (the other dominant mode of heat transfer to the liquid pool [6]). Accurate evaluation of radiative heat transfer can be critical in evaluating fire related problems such as smoke management [2,7,8] and flame inhibition by water sprays [9–14]. To simulate realistic pool fire scenarios, appropriate

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radiation transport models must be incorporated to predict heat feedback to the fuel surface to find liquid vaporization rates.

As pool fire sizes increase, the radiation path length for the flame emission back to the pool surface grows thereby increasing the effect of gasphase absorption inside the flame on radiative heat transfer. Several fire modelers have tried to model the radiation absorption inside the flame and its effects on particularly large pool fires [15-17] including models using NIST's Fire Dynamic Simulator [18] with large-scale eddy simulations of the inherently turbulent buoyant flames. However, a limitation with these modeling efforts is the poor handling of the radiation absorption by the fuel rich core between the flame and the pool surface. This weakness has in part been driven by a lack of radiation absorption data as a function of temperature for typical pool fire fuels such as methanol, toluene, and heptane and in particular, in the oft-used RADCAL database [19]. Studies have worked around this limitation by using a simple gray-gas absorption coefficient inside the pool flame [16,20,21] or by substituting spectral absorption data for a similar fuel (such as CH4 for CH₃OH [15]). However, in general these models have difficulties predicting thermal energy feedback to the fuel surface and the associated mass burning rate for pool fires. Thus, there is motivation to improve the radiation absorption database for gas-phase fuel molecules relevant for pool fires, and several studies over the past decade have contributed to that effort [22-24].

The limited data on spectrally resolved radiation absorption coefficients of fuel species relevant for pool fires has motivated recent research [25,26] to measure fuel radiation absorption coefficients over a range of temperatures (up to 1000 K) that might be expected to exist inside a pool fire flame in the fuel rich core. This research has further explored the importance of spectrally resolved radiation absorption properties in liquid pool fires.

In this study, spectrally resolved absorption coefficients of methanol are measured via FTIR experiments for temperatures up to 1000 K and then used with spectral absorption coefficients for combustion products to solve the 1-D spectrally resolved radiative transport equation along the central axis of a methanol pool fire. Species mole fractions and temperature along the pool fire central axis were obtained from 3-D simulations of FDS utilizing CH₄ absorption coefficient data in place of CH₃OH. The 1-D spectrally resolved calculations were carried out for methanol fires with $d_{\rm pool} = 0.3 \,\mathrm{m}$ and as such were compared with experimental data for similarly sized pools [5]. By exploring alternative simplified radiative absorption models for the fuel species, insight was gained into the selection of radiation models for pool fire simulations.

2. Experimental measurements

2.1. Approach

To provide improved absorption coefficient for methanol, extensive transmission measurements were carried out in a modified FTIRbased rig. Vaporized methanol (99.98% J.T. Baker) infrared spectra were taken over temperatures ranging from 300 to 1000 K. A threestage gas bubbler (sketched in Fig. 1a) in a temperature-controlled water bath and with a bypass N₂ dilution stream was used to supply flows with a range of CH₃OH partial pressures (from 1500 to 3500 ppm for the current study). The N₂-diluted CH₃OH flows were fed to a 32 cm long gas cell in a multi-zone furnace and with ZnSe windows on each end for absorption measurements (as shown in Fig. 1b). For the high temperature measurements, the ZnSe windows were externally cooled to maintain high spectral transmissivity. Further details of the setup are provided elsewhere [25,26].

Control of the methanol partial pressures out of the bubbler was determined by the accuracy of both the temperature measurements in the final bubbler stage (T_{vap}) and of the $P_{vap}-T_{vap}$ relationship where P_{vap} is the fuel vapor pressure. The bubbler was tested to achieve full saturation at the relatively low water bath temperatures in the current study. For P_{vap} , a three-parameter Antoine equation [27] was used

$$P_{\rm vap}(\rm bar) = 1.0 \left(5.20409 - \frac{1581.341}{T_{\rm vap}(\rm K) - 33.50} \right)$$
(1)



Fig. 1. Schematic of experimental apparatus showing (a) three-stage bubbler with dilution flow for controlled liquid fuel concentration and (b) FTIR gas-phase cell with integrated ZnSe window cooling for high-temperature absorption measurements.

In general, based on a 0.5 °C accuracy in the $T_{\rm vap}$ measurement, the concentration uncertainty for the final methanol flow to the measurement cell is calculated to be no more than $\pm 2.5\%$.

Measurements of methanol absorption were taken for 3-5 different partial pressures (usually around 1500, 2300, and 3500 ppm) at each temperature measured (293, 396, 443, 483, 570, 804, and 1000 K). For the measurements, a Mattson Galaxy 7020 FTIR spectrometer was modified such that the IR beam from a globar source passed through the interferometer and was diverted from the internal compartment to the axis of the furnace-housed quartz flow cell. The IR beam passed through the cell and into an external Mercury Cadmium Telluride (MCT) detector (MI0465, Graseby Infrared). Absorption spectra were measured at 1.0 cm^{-1} resolution and averaged over 128 scans.

2.2. Experimental results

Figure 2 shows how the important absorption bands for methanol at room temperature conditions impact blackbody emissions at a range of temperature values. The temperature-dependent normalized blackbody spectral emissive power varies as a function of wavenumber. This figure represents how characteristic bands of methanol impact absorption of blackbody emission. In this regard, measurements show that methanol absorption includes a strong C–O stretching peak centered about 1033 cm⁻¹, a CH₃-bending peak at 1470 cm⁻¹, CH₃-stretching peaks centered at 2981 and 2844 cm⁻¹, bending peaks at 1455 and 1345 cm⁻¹, and an –OH stretching peak at 3680 cm⁻¹.

Figures 3a–d show how the spectral absorption coefficient of the prominent methanol bands vary over the range of temperatures studied. The C–O stretching band, centered around 1033 cm⁻¹, has the most significant impact on absorption of blackbody radiation for lower temperatures



Fig. 2. Measured spectral absorption coefficient of CH₃OH at 293 K and normalized blackbody spectral emissive power for 400, 800, and 1200 K.

 \leq 800 K as suggested by Fig. 2. As temperature increases near a flame zone and the blackbody spectra favors higher and higher frequencies, the broader CH3-bending band, centered around 3000 cm^{-1} , becomes more important for blackbody absorption. Thus, while methanol pool fires generally have shorter fuel rich cores than equivalent diameter heptane and toluene flames [5], the presence of the C-O stretching band in the CH₃OH molecule increases the effective radiative absorption per unit length in the low-temperature regions of the fuel rich core closer to the pool surface. The CH₃-stretching motion, present in all of the fuels mentioned, has a larger impact in regions near the flame where temperatures are high but also where absorption coefficients are significantly reduced as illustrated in Fig. 3.

The presence of C–O bonds in oxygenated fuels such as CH₃OH can have a significant impact on the overall radiative transfer back to the fuel surface in pool fire scenarios. Figure 4 plots the Planck mean absorption coefficient for CH₃OH as function of *T* obtained by integrating the product of the spectral absorption coefficient κ_{ν} and the blackbody Planck function $E_{b\nu}$ over all wavenumbers ν

$$\kappa_{\rm p}(T) = \frac{1}{\sigma T^4} \int_0^\infty \kappa_{\rm v} \frac{C_1 v^3}{\exp\left(\frac{C_2 v}{T}\right) - 1} \mathrm{d}v \tag{2}$$

where C_1 , C_2 and σ are the first and second radiation constants and the Stefan-Boltzmann constant, respectively. κ_{v} is the spectral absorption coefficient. The Planck mean absorption coefficient peaks at 350 K and monotonically decreases with increasing temperature as the strongly absorbing C-O stretch band becomes less significant with the blackbody emission shifts to higher wavenumbers. Figure 4 illustrates qualitatively how radiative absorption by CH₃OH is most significant near the pool surface where the temperature is low and significant radiative emission from the fire is spectrally favorable for absorption. It should be noted that similar calculations for CH₄ yield a $\kappa_{\rm v}$ that is approximately an order of magnitude less than κ_v shown for CH₃OH between 300 and 400 K.

3. Modeling study

3.1. Approach

The effects of the temperature and frequency dependence of fuel radiation absorption coefficients on pool fire heat transfer can be explored by looking at ideal models of pool fires which are adequately simple such that fully resolved spectral and temperature dependency can be explored. For modeling radiative transport for spectral resolutions of 1 cm^{-1} or less, 1-D flame





Fig. 4. Calculated Planck mean absorption coefficient of CH_3OH as a function of blackbody radiation temperature.

models are most manageable. Thus, in the current study, 1-D normal radiative heat transfer along the central axis of a pool fire was used to explore the quantitative effects, in particular, of CH_3OH radiative absorption on heat feedback in pool fires. The 1-D radiation transport along the central pool axis is governed by the general radiative transport equation

$$\frac{\mathrm{d}I_v}{\mathrm{d}z} = -\kappa_v (I_v - I_{bv}) \tag{3}$$

where I_{ν} and $I_{b\nu}$ are the spectral radiation intensity and blackbody spectral emission intensity respectively, and z is height along the axis. To integrate this equation, optical depth τ is introduced as

$$\tau_v = \int_0^z \kappa_v \mathrm{d}z \tag{4}$$

Multiplying both sides of Eq. (3) by $exp(\tau_v)$ and integrating over some unspecified height change leads to the following equation for I_v :

$$I_{v}(\tau_{v}) = I_{v}(0) \exp(-\tau_{v}) + \exp(-\tau_{v})$$
$$\times \int_{0}^{\tau_{v}} I_{bv}(\tau_{v}^{+}) \exp(\tau_{v}^{+}) \mathrm{d}\tau_{v}^{+}$$
(5)

The blackbody source function in Eq. (5) must be integrated numerically. With 1.0 cm^{-1} resolution data, a simple trapezoidal rule was used to determine the integrated radiative heat transfer in Eq. (5). The spectrally resolved radiation transport equation solver was implemented in Matlab, and for a given species and temperature distribution, the radiation transport in a flame environment can be calculated.

Fig. 3. Measured spectral absorption coefficients for CH_3OH for a range of temperatures. Bands are shown that provide significant radiative absorption: (a) C–O stretching band, (b) C–H bending band, (c) C–H stretching band, and (d) O–H stretching band.

To provide a first evaluation of the effects of frequency-resolved and temperature-dependent fuel vapor absorption coefficients, the integrated radiation transport equation (Eq. (5)) was applied in a post-solution manner to simulated methanol pool fire profiles of temperature and species mole fraction derived from NIST's Fire Dynamic Simulator (FDS ver. 4.02 [18]). With its large-eddy simulation approach, FDS can use relatively large grids (in this case $2 \text{ cm} \times 2 \text{ cm} \times 2 \text{ cm}$ grid) with the sub-grid flame models based on mixture fraction to capture flame dynamics without resolving locally steep species and temperature gradients specifically near the flame surface. A finer grid of 1 cm cubes gave similar major species profiles and peak flame temperature (within 20 K) at the same location as the 2 cm cube grid.

4. Results

The species and temperature data along the central axis of a pool fire were tabulated from the mean conditions taken from a FDS simulation of methanol pool fire with $d_{\text{pool}} = 30$ cm. The simulations were run for 60 s at which point the buoyant plume appeared to reach near cyclic behavior, and time-averaged temperature and species profile could be defined for solving the spectrally resolved radiation transport equation (Eq. (3)). The mean temperature and mass fractions for major absorbing species were derived by averaging the FDS simulation results from 50 to 60 s. Figure 5 shows the averaged values along the central axis above the pool surface that were used for the 1-D radiation calculations. The averaged profiles of species and temperature are rather smooth due to the averaging of the transient large-grid LES simulations in FDS. The rapid temperature jump within 2 cm above the pool is very much in agreement with experimental measurements of temperature



Fig. 5. Integrated normal radiation intensity, species mole fraction for major species, and temperature as a function of height for a simulated CH₃OH pool fire with $d_{\text{pool}} = 0.3 \text{ m}$.

profiles in 31 cm methanol pool fires [28]. The average flame height based on methanol conversion reaching 99.9% is 0.74 m is approximately 40% higher than measurements based on luminosity [5]. The discrepancy can be attributed to differences in the definition flame location and to inaccuracies in the FDS combustion models. Nonetheless, the 1-D radiation transport calculations along the central axis of the simulated flame can provide for a basis for comparing models using different fuel absorption coefficient models with the experimental measurements of 30 cm pool fires.

To calculate the radiation intensity normal to the pool surface along the centerline, an appropriate far-field boundary condition must be defined. While for sooting flames, it is often assumed that the soot emissions in the flame can be approximated as a blackbody emission at the flame temperature, such an assumption is not appropriate, and does not work well for the non-luminous and completely non-sooting methanol flames. To this end, for the methanol flames, a flame condition was defined which set a flame temperature at 1400 K based on reviews of past pool fire research on methanol and other fuels [29]. This temperature was set at the location where the methanol conversion reached 99.9% (at a height of 0.74 m). Inputting this high temperature was necessary because the LES simulations of FDS do not resolve the local temperature peak in the flame zone and therefore underpredict the flame emissions. The spectral emission of the flame product species at this high temperature location provided the boundary condition for the radiative transport equations. The averaged temperatures and species concentrations were used for calculating the spectrally resolved absorption coefficient needed in the radiative transport equation which was integrated from the flame location back to the pool surface. For these calculations, a semi-empirical correlation based on a three-parameter fit [25] of the quantum molecular line intensity equation for each wavenumber provided a means for calculating each κ_v as a function of *T*.

Figure 5 shows that H₂O and CO₂ are the principle emitting species near and outside of the actual flame. The methanol mole fraction only becomes significant below a height of 0.3 m (or 0.26 m above the liquid surface). FDS implements a simplistic CO model which assumes that CO is a function of the moles of fuel combusted and therefore indirectly of mixture fraction. This limited model underpredicts CO inside the flame zone and gives CO mole fractions in the ppm level throughout the flame, which is unrealistically low in comparison to gas sampling measurements in experiments ($X_{CO} \sim 0.03$) [30]. However, even measured mole fractions of CO are relatively low compared to H₂O, CO₂, and CH₃OH (inside the flame), and as such it is expected that error in the radiation calculation due to uncertainty in the CO mole fraction will not be significant.

To better appreciate the importance of including methanol absorption into the pool fire calculations, calculations of the fuel emissions intensity (I_{fuel}) as well as the total emissions intensity (I_{tot}) , both integrated over the entire spectrum, are plotted in Fig. 6. As expected, the fraction of radiative emissions back to the pool surface attributable to CH₃OH increases from 0 at a height of 0.55 m and increases to 40% at the pool surface. The increase in CH₃OH concentration combined with the shallow drop in the centerline temperature profile actually leads to an increase in the radiation intensity from the flame towards the fuel surface until 10 cm above the fuel surface. As the temperature drops nearer the fuel surface, the increase in absorption vs. emission causes the radiation intensity to drop for heights <10 cm. For the calculations, I_{tot} at the pool centerline was calculated to be 12,400 W/m² sr at fuel surface, and this value is extremely close to the normal radiation measurements along a pool centerline by Hamins et al. where I_{tot} was measured to be 12,200 $W/m^2 \text{ sr } [5].$

The liquid CH₃OH pool may be transparent to some wavelengths reaching its surface. In general, liquid fuel absorption spectra have broader peaks and additional peaks due to association and higher molecular density. For spectral regions where CH₃OH liquid absorption is small, it is expected that the pool container will absorb that radiation and conduct the heat into the pool, thereby providing an indirect transfer of the radiant heat into the liquid.

Figure 7 plots spectrally resolved radiation intensity at the fuel surface (at height of 0.04 m). The radiation intensity spectrum is plotted with shaded regions showing where the CH₃OH absorption bands lie. Because the simulations suggest that the 3-dimensional flame keeps the fuel



Fig. 6. Integrated normal radiation intensity by all species including CH₃OH, and the ratio of methanol radiation to all species intensity as a function of height for a simulated CH₃OH pool fire with $d_{pool} = 0.3$ m.



Fig. 7. Normal radiation intensity along the flame centerline to the center point of the pool surface for a simulated CH₃OH pool fire with $d_{\text{pool}} = 0.3$ m.

core relatively hot (above 700 K) all the way down to within one grid cell (2 cm) of the pool surface, the spectrally resolved intensity peaks closer to the CH₃-bending band rather than the otherwise stronger C–O stretching band. Exclusion of the fuel absorption from this calculation would increase the overall I_{tot} by approximately 50%.

As many studies include less accurate models of fuel absorption coefficients than the spectrally resolved, temperature-dependent absorption coefficients used in the current study, the question should be asked as to what impact simpler fuel absorption models might have on I_{tot} calculations at the liquid pool surface. Figure 8 compares the calculated radiation intensity as a function of height above the fuel surface for the simulated methanol pool fire using different approaches to radiation absorption modeling of the fuel. The baseline method as described above uses a 1.0 cm^{-1} spectral resolution with temperature



Fig. 8. Comparison of radiation intensity profiles calculated with different approaches to $\kappa_{v,\text{fuel}}$ including temperature-dependent, fixed at ambient temperature, and temperature-dependent with CH₄ values replacing CH₃OH.

dependent CH₃OH absorption coefficients derived from fits to the FTIR experiments. A second approach compared these results to calculations in which the temperature dependence was removed and constant spectrally resolved absorption coefficients for the fuel were used at ambient temperature conditions. This is of interest because ambient temperature optical data is typically available for a variety of fuel species. The results plotted in Fig. 8 show that with the constant low temperature κ_v for the fuel, the methanol pool flame has higher normal radiation intensity closer to the flame where emissions exceed absorption. This can be expected because of the higher value for κ_v at ambient temperature, and the local normal intensity is close to 20% higher at heights between 0.2 and 0.3 m. However, for cooler temperatures near the pool surface, absorption exceeds emission, and the higher ambient temperature κ_{ν} results in a more rapid decline in normal radiation intensity, such that the incident normal $I_{\rm tot}$ at the fuel surface only changes by $\sim 3\%$ (from 12,400 to 12,000 W/cm² sr) in comparison to the temperature-dependent case. This agreement shows that offsetting errors can occur in I_{tot} calculations when using proper spectral bands at a constant temperature if the fuel rich core has significant regions where emission > absorption and where absorption < emission.

A third means of calculating the radiation intensity profile is also shown in Fig. 8, which is based on replacing the CH₃OH temperature-dependent κ_{v} 's with those for CH₄. This is motivated by the fact that some studies have used CH₄ κ_{v} from the RADCAL database [19] as a substitute for fuels in which temperature-dependent κ_v data are not available. Not surprisingly, the use of the CH₄ κ_{v} data underpredicts the emission and subsequent absorption with the former being more significant and resulting in an underprediction of I_{tot} by 20% at the methanol pool surface. Thus, if compromises are to be made in absorption coefficient modeling, it is suggested that relaxations in temperature-dependent data is more acceptable than use of temperature-dependent data without the proper spectral bands. This is particularly true for longer path lengths expected in medium to large pool fires where the spectral band absorption may result in complete loss of radiation intensity as shown in Fig. 7 for the CH₃-bending band.

5. Conclusion

Infrared absorption spectra of methanol were measured using unique high temperature FTIR for temperatures ranging from 300 to 1000 K. As temperature increases, absorption coefficients decrease around band peaks, but the band wings increase due to shifted and broadened rotational transitions. From the radiation absorption coefficients for methanol Planck mean absorption coefficients for blackbody radiation temperatures ranging up to 1400 K were calculated and found to decrease monotonically with increasing temperature above 350 K.

The radiation intensity at the pool surface of 0.3 m diameter methanol pool fire was calculated using 1-D spectrally resolved radiative transport equation with species concentration and temperature profiles derived from averaging transient FDS calculations. The integrated normal radiation at the pool surface center point from the simulations with the spectrally resolved radiation transport agreed extremely well (\sim 3%) with past experimental measurements. Thus, the assumption of an optically thin flame seems to be quite reasonable for the non-sooting methanol flames. The effect of temperature dependency on the absorption coefficient on the radiation intensity at the fuel surface was also investigated by comparing calculations with temperature-dependent κ_{v} and with constant ambient κ_{v} . Although the variation in κ_v with temperature was large over the range of temperatures in the fuel-rich core, the difference in I_{tot} at the methanol pool surface was small ($\sim 2\%$) due to compensating errors between the increased emissions and absorption with the higher κ_v at ambient temperature. All the same, the favorable comparison with experiments suggests the value in careful experimental tabulation of fuel-radiation absorption coefficients for use in full-scale simulations for predicting the dominant heat feedback mechanism of radiation in pool fire scenarios.

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