A diffusion-based approximate model for radiation heat transfer in a solar thermochemical reactor

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Abstract

An approximate method for fast calculations of the radiation heat transfer in a solar thermochemical reactor cavity is proposed. The two-step method with separate calculations for solar and thermal radiation is based on the diffusion approach. The usual $P_1$ approximation is generalized by use of an equivalent radiation diffusion coefficient for the optically thin central part of the cavity. The resulting boundary-value problems are solved using the finite element algorithm. The accuracy of the model is assessed by comparing the results to those obtained by a pathlength-based Monte Carlo simulation. The applicability of the proposed model is demonstrated by performing calculations for an example problem which incorporates a range of parameters typical for a solar chemical reactor and spectral radiative properties of polydisperse zinc oxide particles.

Nomenclature

- $a$: particle radius, m
- $A$: surface area, m$^2$
- $A, B$: parameters of the particle size distribution function
- $D$: radiation diffusion coefficient, m
- $f_v$: particle volume fraction
- $I\lambda$: direction-integrated spectral radiation intensity, W\text{-m}^{-3}\text{\mu m}^{-1}
- $L$: cavity length, m
- $I_b$: spectral radiation intensity, W\text{-m}^{-2}\text{sr}^{-1}\text{\mu m}^{-1}
- $k$: index of absorption
- $l$: path length, m
- $m$: complex index of refraction
- $n$: index of refraction
- $n_{MC}$: number of MC sub-samples
- $n_{ray}$: total number of rays in MC
- $Q$: efficiency factor
- $q$: thermal power, W
- $q'$: integral radiation flux, W\text{-m}^{-2}
- $r$: radial coordinate, m
- $R$: cavity radius, m
- $R_1$: aperture radius, m
- $R_2$: radius of the cloud-bed interface, m
- $T$: temperature, K
- $V$: volume, m$^3$
- $x$: diffraction parameter
- $z$: axial coordinate, m

Greek symbols

- $\beta$: extinction coefficient, m$^{-1}$
- $\varepsilon$: total hemispherical emissivity
- $\theta$: polar angle, rad
- $\kappa$: absorption coefficient, m$^{-1}$
- $\lambda$: wavelength, $\mu$m
- $\nu$: number of degrees of freedom
- $\sigma$: Stefan–Boltzmann constant
- $\sigma_s$: scattering coefficient, m$^{-1}$
- $\Phi$: scattering phase function
- $\varphi$: azimuthal angle, rad

Subscripts

- $a$: absorption
- $b$: blackbody
- back: back wall
1. Introduction

Concentrated solar radiation is used as the energy source of high-temperature heat for advanced thermochemical processing [1]. It offers several advantages over the combustion-based technology. Energy is efficiently transferred via direct irradiation to the reaction site, bypassing the limitations imposed by indirect heat transfer through heat exchangers in external combustion, or eliminating the quality degradation of chemical products by combustion gases and soot in internal combustion. The emission of greenhouse gases is avoided, and in the case of the fuel production or upgrade, the final product contains chemically stored solar energy. Example applications include the thermal decomposition of limestone, the thermal reduction of metal oxides, the thermal cracking of natural gas, and the thermal gasification of coal. In particular, solar high-flux irradiation is used for the thermal dissociation of zinc oxide

\[ 2\text{ZnO}(s) \rightarrow 2\text{Zn}(g) + \text{O}_2(g) \]  

(1)

at temperatures above 2100 K [2]. The importance of this reaction relies in the variety of applications of Zn as an energy carrier, e.g. its hydrolysis for hydrogen production. The solar reactor features a rotating cavity-receiver lined with micron-size zinc oxide particles that are directly irradiated and decomposed. A complete description of the combined heat transfer in the reactor includes the complex spectral description of thermal radiation by walls, particles, and of the incoming solar flux for regions by accounting for varying particle concentration within the receiver.

Previous pertinent studies of radiative heat transfer within particle suspensions exposed to concentrated solar radiation include steady-state models based on the discrete ordinates method [3], on the six-flux method [4], and on the Monte-Carlo (MC) method [5]. Radiative heat transfer analysis in participating media undergoing thermochemical transformation under direct irradiation was performed by a steady-state model based on MC [6], and transient models based on the Rosseland approximation [7, 8] and on MC [9]. Iterative or transient computations employing the general MC technique require time-consuming full ray-tracing run for each iteration or time step.

This paper presents an approximate method for fast calculations of the radiative heat transfer in a solar thermochemical reactor cavity containing ZnO particles of spatially varying size and concentration. The method is based on the diffusion approach (DA) [10, 11]. The accuracy of the model is assessed by comparing the results to those obtained by a pathlength-based MC simulation [12]. The applicability of the proposed model is demonstrated by performing calculations for an example problem which incorporates a range of parameters typical for a solar chemical reactor and spectral radiative properties of ZnO particles.

2. Problem statement

A solar thermochemical reactor for the thermal dissociation of semi-transparent ZnO particles under concentrated solar irradiation is considered [13]. The reactor features a rotating cylindrical cavity with a window aperture at its front wall, while ZnO particles are supplied by
the axial feeding system, operating cyclically. Small particles fall irregularly by gravity and inertial forces to the lateral wall while being heated and decomposed into the gaseous products. The geometry of the model problem is depicted in figure 1. Three axisymmetric regions within the cavity are distinguished: (1) the non-participating empty central part; (2) the particle cloud of small ZnO particles; and (3) the particle bed at the lateral wall.

Fig. 1 – Scheme of a thermochemical reactor cavity.

The particle average radius \( a_{32} = 10 \mu m \) and volume fraction \( f_v = 10^{-3} \) are assumed in the cloud and \( a_{32} = 500 \mu m, f_v = 10^{-1} \) are assumed in the particle bed. The value \( a_{32} \) is defined as follows:

\[
a_{32} = \frac{\int_0^x a F(a) da}{\int_0^x a^2 F(a) da}
\]

where \( F(a) \) is the size distribution function.

The cavity walls are assumed to be gray and diffuse with hemispherical emissivity of \( \varepsilon_w = 0.8 \) and at uniform temperature 1600 K. The incoming integral solar flux \( q_{in} = 2.5 \text{ MW/m}^2 \) is assumed diffuse and uniformly distributed over the aperture, but the model can readily be extended to include directional distributions of incident solar radiation as discussed later. Its spectral distribution is approximated by that of a blackbody at \( T_{in} = 5780 \text{ K} \). The particle temperature in the cloud is assumed constant and equal to 2200 K. In the particle bed, we assume the linear radial dependence of particle temperature from 2000K to the wall temperature.

3. Radiative properties

Radiative properties of the participating medium within the cavity are derived by neglecting the radiation absorption by gaseous phase, consisting of \( \text{O}_2, \text{Zn} \) and \( \text{Ar} \) at atmospheric pressure. The particle radiative properties are obtained based on the optical constants of ZnO, the local particle volume fraction \( f_v \), and the particle size distribution \( F(a) \). The index of refraction is assumed constant \( n_a = 1.9 \). The index of absorption is obtained by approximating the data from [15] as
where \(k_0 = 0.5\), \(\lambda_i = 0.37\,\mu\text{m}\), \(\gamma = 120\,\mu\text{m}^2\), and \(k_1 = 10^{-4}\).

Variation of the absorption index with the parameter \(k_1\) is shown in figure 2. Plotted are also the values obtained from the theoretical model [15]. While the refractive index is widely available in the literature [15–18], the infrared absorption index of ZnO is not reported, and relation (3) is only an assumption in the range \(\lambda > 0.8\,\mu\text{m}\). Since the long-wave index of absorption strongly varies with the parameter \(k_1\), it may lead to a significant error in the infrared absorption and emission by particles.

The transport approximation of the scattering phase function is employed in this paper. This enables to simplify the formulation of a radiative transfer problem. As a result, only the efficiency factor of absorption and the transport efficiency factor of extinction are required. This approach is known to be sufficiently accurate for radiative transfer calculations in many applications [10, 11].

The efficiency factors for absorption, transport scattering and transport extinction of a single weakly-absorbing particle are calculated as [14]

\[
Q_{\text{a,\lambda}} = \frac{4n_i}{(n_i + 1)^2} [1 - \exp(-4k_\lambda x)] ,
\quad Q_{\text{a,ir,\lambda}} = \left\{ \begin{array}{ll}
C \zeta, & \zeta \leq 1 \\
C/\zeta, & \zeta > 1
\end{array} \right.
\quad Q_{\text{ext,ir,\lambda}} = Q_{\text{a,\lambda}} + Q_{\text{a,ir,\lambda}}
\]

where \(C = 1.5n_i (n_i - 1) \exp(-15k_\lambda)\), \(\gamma = 1.4 \exp(-80k_\lambda)\), \(\zeta = 0.4(n_i - 1)x\) and \(x = 2\pi a/\lambda\). The absorption and transport scattering efficiency factors of ZnO particles for particle radii \(a = 10, 100\) and \(500 \,\mu\text{m}\) and \(k_1 = 10^{-4}\) are shown in figure 3. Also plotted are curves for the efficiency factors obtained from the Mie theory. The approximation of the absorption efficiency factor is accurate for both small and large particles over the whole spectral range. In contrast, the agreement between the approximated and Mie transport efficiency factors of scattering is good for large particles but overestimates the scattering by
Fig. 3 – Radiative characteristics of ZnO particles for particle radii $a$ (1) – 10, (2) – 100 and (3) – 500 $\mu$m, calculated by using approximation (4), and by the Mie theory.

small particles in the long-wave range. The latter error is expected to have minor impact on the radiative transfer in the model problem due to insignificant role of the long-wave radiation scattering by cloud particles.

Absorption, transport scattering and transport extinction coefficients of polydisperse particles are obtained from [10]

$$
\kappa_\lambda, \sigma_{s,tr,\lambda}, \beta_{tr,\lambda} = 0.75 f_v \int_0^\infty \left\{ Q_{a,\lambda}, Q_{s,tr,\lambda}, Q_{ext,\lambda} \right\} a^2 F(a) da \int_0^\infty a^3 F(a) da \quad (5)
$$

where $f_v$ is the particle volume fraction. In the monodisperse approximation based on the average particle radius $a_{32}$, the absorption, scattering and extinction coefficients are given by

$$
\kappa_\lambda, \sigma_{s,tr,\lambda}, \beta_{tr,\lambda} = 0.75 f_v \left\{ Q_{a,\lambda}, Q_{s,tr,\lambda}, Q_{ext,\lambda} \right\} / a_{32} \quad (6)
$$

where the efficiency factors are determined for $a = a_{32}$.

Fig. 4 – Absorption and transport scattering coefficients: 1 – $a_{32} = 10$, 2 – 500 $\mu$m.
Figure 4 shows the spectral radiation transfer coefficients of ZnO particles for \( k_1 = 10^{-4} \). The curves obtained in the monodisperse approximation agree very well with those for the selected polydisperse systems (\( F(a) = \exp[-(a-a_m)^2/(2\sigma^2)] \)) for both \( a_m = 10 \text{ \mu m} \) and \( 500 \text{ \mu m} \) in the considered spectral region. Therefore, the monodisperse approximation is selected for further analysis. The results of figure 4 also exemplify the predominance of absorption and scattering by large particles.

4. Reference Monte Carlo solution

The pathlength MC with ray redirection is applied to solve for the radiative exchange in the cavity [12]. The net radiative flux to a wall element \( dA \) and the divergence of the radiative flux within an elementary volume \( dV \) of the participating medium are calculated as

\[
q_{r,w} = \frac{\sum_{k_w} \sigma_w q_{ray,k_w}}{dA_w} - \sigma_w \sigma_T^w \quad (7)
\]

\[
\nabla \cdot q_{r,p} = 4\kappa_{p,p} T_p^4 \frac{\sum_{k_p} q_{ray,k_p} K_{k,p} d_{l_p}}{dV_p} \quad (8)
\]

where \( k_w \) and \( k_p \) designate rays reflected by the wall element \( dA_w \) and traversing the medium volume \( dV_p \), respectively. \( q_{ray,k_p} \) is power of \( k_p \)'th ray entering volume \( dV_p \) and \( d_{l_p} \) is the path length of \( k_p \)'th ray within \( dV_p \). The probability density functions for the azimuthal and polar angles \( \phi \) and \( \theta \), and the wavelength \( \lambda \) for emission/scattering by the non-gray medium, and for emission/reflection by gray-diffuse walls are employed in the MC code [11, 12]. Isotropic scattering is assumed to make the MC procedure conforming to the transport approximation. The finite volume technique is applied to obtain the discrete form of equations Eqs. (7) and (8) on the structured cylindrical grid.

5. Diffusion approximation

In the diffusion approximation, the spectral radiative flux is adopted in the following form [10]

\[
q_{r,\lambda} = -D_\lambda \nabla G_\lambda , \quad G_\lambda = \int_0^{4\pi} I_\lambda d\Omega \quad (9)
\]

where \( D_\lambda \) is the radiation diffusion coefficient, proportional to the mean free path of radiation. Substituting Eq. (9) into the direction-integrated equation of radiative transfer leads to the boundary-value problem for the direction-integrated spectral radiative intensity \( G_\lambda \) [10]. In the diffusion approach presented here, it is assumed that radiation heat losses from the cavity are determined mainly by thermal radiation of particles and walls, whereas the contribution of the scattered solar radiation is negligible. This enables one to split the solution into two steps. In the first step, the radiative exchange in the cavity is solved for the thermal radiation emitted by the walls and particles with the inclusion of the central (non-participating) part of the medium region while omitting solar radiation. The corresponding boundary-value problem is formulated as follows:
The usual form of the diffusion coefficient $D_{\alpha} = \frac{1}{3\beta_{u,\alpha}}$, which corresponds to the $P_1$ approximation [10, 11], is used for the particulate medium. However, the medium in the central part of the computational region ($r < R_i$) is not participating and this relation cannot be used for $\beta_{u,\alpha} = 0$. Instead, a constant value of the radiation diffusion coefficient $D_{\alpha} = \frac{2R_iH}{(2R_i + H)}$ is employed for $r < R_i$ by recalling the physical meaning of $D_{\alpha}$. The model calculations showed that the results are not sensitive to the choice of this expression.

In the second step of the solution, the reflection of solar radiation from the particle cloud and from the central part of the back wall is neglected. Hence, the spectral solar radiation fluxes to the cloud and to the central part of the back wall are given by [11]

$$q_{\lambda,c} = \frac{q_m}{\sigma T_i^4} \pi I_{2b} (T_i) \left( \frac{Z^2 + 0.5}{\sqrt{Z^2 + 1}} - Z \right), \quad Z = z/(2R_i)$$

$$q_{\lambda,w,back} = 0.5 \frac{q_m}{\sigma T_i^4} \pi I_{2b} (T_i) \varepsilon_w \left( X - \sqrt{X^2 - 4} \right), \quad X = 2 + \left( L/R_i \right)^2$$

Note that equations (14) and (15) are valid only for the diffuse incident solar flux. For non-diffuse distribution of incident solar radiation, the fluxes to the cloud and to the central part of the back wall must be obtained by preserving the directional character of the incident solar radiation, e.g. by using MC. However, the directional character of the incident solar flux will be lost in the participating medium.

Thus, the boundary-value problem for the second step reduces to the computational region including the particle cloud and the bed only. The thermal radiation from particles and walls is omitted, but the incoming solar flux is included in the boundary condition at the cloud surface. This is formulated as

$$-\nabla (D_{\lambda} \nabla G_{\lambda}) + \kappa_{\lambda} G_{\lambda} = 0$$

$$-D_{\lambda} \hat{n}_w \cdot \nabla G_{\lambda,w} = \gamma_w \left[ G_{\lambda,w} - S_{\lambda,w} \right]$$

$$S_{\lambda,w} = \begin{cases} 4q_{\lambda,c} & \text{for } 0 \leq z < L \text{ and } r = R_i \\ 0 & \text{at the cavity walls} \end{cases}$$

Due to the linearity of the problem, the final solution including radiative flux and its divergence is obtained as a sum of the solutions for the two steps.
The boundary-value problems (10)–(13) and (16)–(18) are solved by the finite element method on the structured triangular grid [10].

6. Results

Numerical computations were performed for the model problem using both MC and DA. Figure 5 shows the integral radiative heat flux to the front (including the aperture) and back walls of the cavity. The agreement between the MC and DA is better for the back wall than for the front wall. The MC and DA profiles converge at $r/R \to 1$.

Axial profiles of the radiative fluxes are shown in figure 6. The best agreement between the MC and DA results is achieved at the lateral wall, where the profiles are almost identical. In contrast, the maximum differences are found at the cloud boundary.

The source of errors of the DA method was studied by performing additional calculations for a case with the solar flux set to zero. It was found that the overestimation of the axial radiative flux originates from the overestimated fluxes due to solar radiation. The radial radiative flux resulting from the incoming solar radiation is modeled properly by DA at selected locations for both the normal case and the case with no solar flux.

The quality of both MC and DA results increases with increasing $r/R$ ratio. While this effect is explained for MC by decreasing size of the control elements with decreasing radius, it is caused by the FEM discretisation in the DA approach. The selected resolution of the unstructured triangular grid in the DA method also leads to oscillations in the curves shown in figures 5–7. The accuracy of the MC results was determined by calculating the $P = 95\%$ confidence intervals $\bar{q} \pm t_{\nu, 0.05} S_{\bar{q}} / \sqrt{n_{\text{MC}}}$ for $\nu = n_{\text{MC}} - 1 = 9$ degrees of freedom. The absolute confidence interval for the radiative flux was maximum at the aperture ($r/R = 0.0167, \dot{q}_{r,w} = -1636.2 \pm 63.5 \text{ kW/m}^2$), whereas for its divergence it was maximum for the bed ($r/R = 0.95, z/L = 0.25, \nabla \cdot \dot{q}_r = 6776.5 \pm 1835.1 \text{ kW/m}^3$).
A typical MC simulation was performed for the total number of rays $n_{ray} = 10^7$ and took about 5000 s on a PC with a Pentium 4 3.2 GHz processor. In contrast, a typical DA simulation lasted less than 3 s. The latter demonstrates a significant time reduction of the suggested approach, and therefore makes the method attractive for solving transient combined heat transfer problems.

The DA model, including the approximations for the radiative characteristics of particles, is convenient for studying the effect of particle parameters on radiation heat transfer in the solar thermochemical reactor. For example, the role of the infrared absorption index of zinc oxide is illustrated in figure 7. Interestingly, radiation absorption in the particle bed is approximately 50% greater in the case of $k_1=10^{-3}$ that in the baseline variant with $k_1=10^{-4}$. This high sensitivity of the results to the infrared absorption index of zinc oxide shows the importance of an experimental validation of the ZnO absorption spectrum in the near infrared.

7. Conclusions

An approximate method based on the diffusion approach for fast calculations of the radiation heat transfer in a solar thermochemical reactor cavity has been presented. The method comprises separate calculations for the incoming solar flux and thermal radiation emitted by the particles and cavity walls. The calculations are based on $P_1$ approximation and the finite element method, and have been performed for a medium with a realistic strong radial variation of the radiative properties due to the varying particle concentration and diameter. The numerical validation of the diffusion approximation method by the reference Monte Carlo technique shows a good agreement of the results, in particular for the radial radiative flux. At the same time, the MC simulation is recommended for an intermediate control of the DA results for typical ranges of the problem parameters.

An approximate form of the absorption and transport scattering efficiency factors for semitransparent ZnO particles is proposed and validated by comparing them with the exact values obtained from the Mie theory. A comparison of radiation coefficients for the mono- and polydisperse medium shows that monodisperse approximation can be employed. The approximate relations for the main absorption and scattering characteristics of particles and the monodisperse approximation lead to additional significant simplification of the calculations.
The method presented enables one a significant reduction of the computational time in calculations for combined transient heat transfer problems, when the radiation field has to be calculated at each time step of numerical solution. It makes possible the implementation of the complete spectral radiation transfer calculations in problem-oriented CFD codes for the complex analysis of the solar thermochemical reactors.

The parameter study performed by using the approximate model shows the importance of the experimental determination of the zinc oxide near-infrared absorption spectrum, which will enable one to improve the numerically predicted heat generation rate in the particle bed.

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