HEAT AND MASS TRANSFER MODEL OF A PACKED-BED REACTOR FOR SOLAR THERMOCHEMICAL CO₂ CAPTURE

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ABSTRACT

A 1 kWth dual-cavity solar thermochemical reactor concept is proposed to capture carbon dioxide via the calcium oxide based calcination–carbonation cycle. The reactor is oriented beam-up wherein concentrated solar energy from a heliostat field enters an aperture located at the bottom of the reactor. In the endothermic calcination step, concentrated solar radiation is captured by the inner cavity and transferred by conduction through a diathermal cavity wall to the particulate CaCO₃ medium located in the annular reaction zone. The liberated CO₂ is removed from the reactor to external storage. In the exothermic carbonation step, a CO₂-containing gas flows through a bed of CaO particles in the reaction zone, forming CaCO₃, while CO₂-depleted gas leaves the system. The reactor design is refined using a numerical heat and fluid flow model for the calcination step. The Monte Carlo ray-tracing method is employed to solve for radiative exchange in the inner cavity, coupled with a computational fluid dynamics analysis to solve the mass, momentum, and energy equations in the concentric reaction zone modeled as a gas-saturated porous medium consisting of optically large semitransparent particles. The cavity diameter and length-to-diameter ratio are varied to study their effects on pressure drop, temperature distribution, and heat transfer in the reactor. Increasing the cavity diameter and length-to-diameter ratio decreases the radial temperature gradients across the cavity wall and within the reaction zone. However, it also results in increased pressure drop and reduced heat transfer to the reaction zone.

KEY WORDS: Solar energy, Carbon emission mitigation, Radiation, Chemical reactor modeling, Carbonation–calcination cycle

1. INTRODUCTION

In recent years, it has become apparent that anthropogenic greenhouse gas emissions are contributing to global climate change [1]. Carbon dioxide (CO₂) emissions make up 77% of all greenhouse gas emissions worldwide [2]. In addition, CO₂ is used in many applications, including those in the food and beverage, oil recovery, and manufacturing industries [3–4]. A system to capture CO₂ from dilute sources and turn it into a concentrated stream promises to simultaneously combat climate change and produce an important commodity. One proposed method for CO₂ capture is the calcium oxide (CaO) based carbonation–calcination cycle:

\[
\begin{align*}
\text{Carbonation (exothermic)}: & \quad \text{CaO} + \text{CO}_2 \rightarrow \text{CaCO}_3, \quad \Delta h_{298K}^{0} = -178 \text{ kJ mol}^{-1} \\
\text{Calcination (endothermic)}: & \quad \text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2, \quad \Delta h_{298K}^{0} = 178 \text{ kJ mol}^{-1}
\end{align*}
\]

The exothermic carbonation step proceeds at low temperatures, and the endothermic calcination step becomes thermodynamically favorable at temperatures above 1150 K at atmospheric pressure. Advantages of this cycle include operation at atmospheric pressure and the use of low-cost, naturally available sorbent [5–6].
The cycle (1a)–(1b) has been extensively studied using combustion as the process heat for calcination. Shimizu et al. performed a thermodynamic analysis on a pair of fluidized beds capturing CO₂ from a 1000 MW air fired power plant [7]. Plant efficiency with heat recovery and CO₂ capture was 33.4% compared to 32% for an oxygen fired plant. Abanades et al. experimentally demonstrated that an 18 kW pilot scale dual fluidized bed setup was able to effectively capture CO₂ at carbonation temperatures near 650°C [8]. Rodriguez et al. compared three pilot scale CO₂ capture plants with power of 10–75 kW [9]. Each plant used a different fuel and limestone source, but all plants achieved over 70% CO₂ capture efficiency.

Because of the high temperature required for the calcination reaction, concentrated solar energy could be substituted for combustion as process heat. Nikulshina et al. performed thermodynamic analyses, chemical kinetics experiments, and fluidized bed experimental studies of a solar-driven, CaO-based cycle for CO₂ capture[10–13]. The fluidized bed experiments, which were in a quartz tube reactor, demonstrated complete CO₂ removal in the carbonation step. Meier et al. studied the calcination step of the cycle in two different 10 kW solar rotary kiln reactors [14–15]. One reactor was directly irradiated and the other indirectly irradiated. The directly irradiated reactor achieved solar-to-chemical conversion efficiency of up to 20%, and the indirectly irradiated reactor achieved efficiencies of up to 38%.

Particle-level numerical modeling of the CaO-based CO₂ capture process has focused on the chemical reactions involved in the process. Ebner and Lipiński modeled a CaCO₃ particle undergoing solar-driven calcination [16]. The inclusion of radiation heat transfer significantly decreased the time to reaction completion. Stendardo and Foscolo modeled the carbonation reaction in a particle with coupled chemical kinetics and mass transfer [17]. Diffusion through the CaO product layer was also taken into account, and the model compared well with experimental data.

A few reactor-level modeling studies of solar-driven calciners have been published. Meier investigated a 1.5 MW falling particle reactor directly irradiated by concentrated sunlight [18]. Fluid mechanics and heat transfer, including radiation, were modeled while neglecting chemical reactions. The study identified a need for mechanical components to slow particle acceleration in the curtain and recommended using particles greater than 300 μm diameter due to entrainment of smaller particles into the gas flow. Fidaros et al. studied the effect of thermal input, feed rate, rotational speed, and particle diameter on the efficiency of a 10 kW rotary kiln reactor [19]. Increasing the feed rate and particle size resulted in a higher degree of calcination. The rotational speed and solar energy input did not have much effect on the degree of calcination.

This paper describes a numerical model of a beam up reactor concept to be used for solar CO₂ capture. The reactor geometry and gas inlet temperature are varied to determine their effects on the temperature and velocity profiles within the reactor and on the work required to pump gases through the reaction zone.

2. PROBLEM STATEMENT

The proposed reactor concept is shown in Fig. 1. It consists of two concentric vertical cylindrical cavities. Incident concentrated solar irradiation enters the inner cavity through a windowless aperture at the bottom of the reactor. This beam up configuration has two major advantages: reduction of convective losses through the open aperture and improved optical efficiency compared to beam down due to fewer reflection [20]. Radiation absorbed at the cavity wall is transferred by conduction to the annular space formed by the two cylindrical walls. The annular space is filled with a reacting packed bed of CaCO₃ particles. It is a batch process in which each reaction is consecutively performed. The particles remain inside the reactor throughout the process as the gas species are changed and solar input is switched on and off to drive each reaction. In an industrial scale version of this reactor, the calcination step would be performed during the day and the carbonation reaction performed at night. Gas enters the reactor via inlets into a plenum before passing through a distributor plate into the reaction zone. The gas flows through the annular reaction zone and exits the reactor through outlets at the top.
Fig. 1 Reactor concept for batch mode solar thermochemical CO₂ capture (left: side cross section, top right: top trimetric view, bottom right: bottom trimetric view)

The reaction zone is a packed bed with a particle volume fraction, \( f_{v,s} \), of 0.7. The dimensions of the reactor and the temperature of the gases entering the reactor are varied to study their effects on the temperature and velocity distributions in the reaction zone. The base case values and the ranges of parameters investigated in this study are shown in Table 1. In this study, the reaction zone volume is held constant as the cavity height and diameter are varied. The maximum volume of the reaction zone is calculated by assuming a particle volume fraction of \( f_{v,s} = 0.1 \) to allow for reactor operation as a fluidized bed, a solar input power \( Q_{solar} = 1 \text{ kW} \), a solar-to-chemical conversion efficiency \( \eta = \frac{M_{\text{CaO}} \Delta H_{\text{calc}}^{\text{f}}}{M_{\text{CaO}} Q_{solar}} = 0.35 \), and a calcination time of 30 minutes. The particle volume is assumed to be unchanged during the carbonation–calcination process. Using these assumptions, the mass of CaO produced by calcination is 196.5 g. The reaction zone volume is then given by \( V_t = \frac{M_{\text{CaO}}}{P_{\text{CaO}} f_{v,s}} = 586.5 \text{ cm}^3 \).

This is the anticipated maximum reaction zone volume. For fixed nominal reactor thermal power and decreasing reacting efficiency, the reaction zone volume decreases for a given reaction time.

The solar radiative flux entering the cavity is assumed to be uniformly distributed within a cone angle of 45° with a total power of 1 kW and a concentration ratio of 1000.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Unit</th>
<th>Baseline value</th>
<th>Value range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cavity radius</td>
<td>( r_{cav} )</td>
<td>cm</td>
<td>3</td>
<td>3–5</td>
</tr>
<tr>
<td>Length-to-radius ratio</td>
<td>( H/r_{cav} )</td>
<td>–</td>
<td>4</td>
<td>4–6</td>
</tr>
<tr>
<td>Heat loss</td>
<td>( q_{wall} )</td>
<td>–</td>
<td>0</td>
<td>0–0.2</td>
</tr>
</tbody>
</table>
3. ANALYSIS

A cross section of the three dimensional computational domain is shown in Fig. 2. For simplicity, only the reaction zone and reactor cavity are modeled. The cavity height is $H$, $r_{cav}$ is the inner radius of the cavity, $r_1$ is the outer radius of the cavity and the inner radius of the reaction zone, and $r_2$ is the outer radius of the reaction zone. The reaction zone is modeled as a homogeneous, radiatively-participating porous medium with local thermal equilibrium between the gas ($CO_2$) and solid ($CaCO_3$) phases. The cavity wall is modeled as an opaque, isotropic solid. The model is solved until steady state is reached. External body forces including gravity are neglected.

3.1. Mass, Momentum, and Energy Conservation

The heat and mass transfer model is based on the mass, momentum and energy conservation equations [21]. Using the above assumptions and applying to the packed bed region, these equations reduce to:

Continuity:

$$\nabla \cdot (\rho \mathbf{v}) = 0$$

(2)

Momentum:

$$\nabla \cdot (\rho \mathbf{v} \mathbf{v}) = -\nabla p + \nabla \cdot (\mathbf{\tau}) + {\bar{S}}$$

(3)

Energy:

$$\nabla \left[ \mathbf{v} (\rho E_i + p) \right] = -\nabla \cdot {\bar{q}}_{rad} + \nabla \left[ k_{eff} \nabla T + (\mathbf{\tau} \cdot \mathbf{v}) \right]$$

(4)

where $\rho$ is density, $\mathbf{v}$ is the velocity vector, $\mathbf{S}$ is a momentum source term, $\mathbf{\tau}$ is the shear stress tensor, $p$ is the static pressure, $E$ is the internal plus kinetic energy and is equal to $h = \frac{P}{\rho} + \frac{v^2}{2}$, where $v$ is the fluid speed, $q_{rad}$ is the internal radiative heat flux, the effective conductivity is given by the homogenous model,
\[ k_{\text{eff}} = (1 - f_{v,s})k_t + f_{v,s}k_s, \]  
\( T \) is temperature, \( h \) is enthalpy, and the subscripts \( f \) and \( s \) indicate fluid and solid, respectively.

For a homogeneous porous medium, the source term in Eq. (3) accounts for viscous and inertial losses and is calculated with:

\[
\bar{S} = -\left( \frac{\mu}{K} \bar{v} + \frac{1}{2} C_f \rho |\bar{v}| \bar{v} \right) \tag{5}
\]

where \( K \) is the permeability and \( C_f \) is the Forchheimer coefficient of the porous zone. The Ergun equation is used to derive \( K \) and \( C_f \) for the packed bed, resulting in 
\[
\frac{1}{K} = \frac{150 f_{v,s}^2}{D_p (1 - f_{v,s})^3} \quad \text{and} \quad C_f = \frac{3.5 f_{v,s}}{D_p (1 - f_{v,s})^3} \tag{22}.\]

In this study, \( f_{v,s} = 0.7 \) and \( D_p = 100 \, \mu m \). The typical range of solid volume fraction values for monodisperse, randomly packed spheres is \( 0.56 - 0.64 \) with a maximum of 0.74 for cubic or hexagonal close packed spheres [23]. A higher value was chosen as, in reality, the particles will not be completely spherical or monodisperse, resulting in increased solid volume fraction. In the silicon carbide cavity wall region, the energy equation reduces to:

\[
-k_{\text{SiC}} \nabla^2 T = 0 \tag{6}
\]

where \( k_{\text{SiC}} \) is the thermal conductivity of the cavity wall.

### 3.2. Radiative Heat Transfer

The divergence of the radiative heat flux in the packed bed is found by solving the radiative transfer equation, which for a grey medium reduces to [24]:

\[
\nabla \cdot \bar{q}_{\text{rad}}^r = \kappa \left( 4 \sigma T^4 - \int_{\lambda=0}^{\infty} \int_{0}^{4\pi} I_{\lambda} \hat{s} \, d\Omega \right) \tag{7}
\]

For the range of dimensions and radiative properties of the reactive medium considered in this study (see Table 2 and section 4), the minimum optical thickness, \( \tau \), is over 20,000 and the packed bed medium is optically thick. Consequently, the Rosseland diffusion approximation is employed to model the internal radiative heat transfer in the packed bed. The \( \text{CO}_2 \) is assumed to be radiatively nonparticipating, and \( \text{CaCO}_3 \) particles are assumed to be absorbing and independently scattering. Thus, in Eq. (4) the radiative heat flux can be approximated by 
\[
\bar{q}_{\text{rad}}^r = -k_{\text{rad}} \nabla T, \quad \text{where} \quad k_{\text{rad}} = \frac{16 \pi^2 n^2 \sigma T^5}{3 (\kappa + \sigma_s)}, \quad n \text{ is the real part of the refractive index of the host medium,} \quad \kappa \text{ is the absorption coefficient, and} \quad \sigma_s \text{ is the scattering coefficient. The scattering and absorption coefficients are calculated assuming independent scattering and using Mie theory with a characteristic particle diameter of 100 \( \mu m \) [25]. The spectral values of the complex refractive index, } m, \text{ of } \text{CaCO}_3 \text{ are used to obtain } \kappa_\lambda \text{ and } \sigma_{s,\lambda} \text{ and then since the values are nearly constant across the spectrum, } \kappa_\lambda \text{ and } \sigma_{s,\lambda} \text{ are algebraically averaged over the spectrum to obtain } \kappa \text{ and } \sigma_s.\]

### 3.3. Boundary Conditions

The boundary conditions are

- For the continuity equation:
\[ \rho_i \left( \mathbf{v} \cdot \hat{k} \right) |_{r < r_2, z = z_{bottom}} = \rho_i u_{z, in} \]  

(8)

- For the momentum equation:

\[ p |_{r < r_2, z = z_{top}} = 0 \]  

(9)

\[ u_r |_{r = r_1, z = z_{bottom}} = u_r |_{r = r_2, z = z_{top}} = u_z |_{r = r_1, z = z_{bottom}} = u_z |_{r = r_2, z = z_{top}} = 0 \]  

(10)

- For the energy equation:

\[ T |_{r < r_2, z = z_{bottom}} = T_{in} \]  

(11)

\[ \frac{\partial T}{\partial z} \bigg|_{0 < r < r_1, z = 0} = 0 \]  

(12)

\[ -k_{SiC} \frac{\partial T}{\partial r} \bigg|_{r = r_{cav}, z \leq z_{H}} = -k_{SiC} \frac{\partial T}{\partial z} \bigg|_{0 < r < r_{cav}, z = H} = -k_{SiC} \frac{\partial T}{\partial z} \bigg|_{0 < r < r_{cav}, z = z_{top}} = q_{rad} \]  

(13)

\[ k_{SiC} \frac{\partial T}{\partial r} \bigg|_{r = r_1, z \leq z_{top}} = \left( k_{eff} + k_{rad} \right) \frac{\partial T}{\partial r} \bigg|_{r = r_1, z \leq z_{top}} \]  

(14)

\[ -\left( k_{eff} + k_{rad} \right) \frac{\partial T}{\partial r} \bigg|_{r = r_1, z \leq z_{top}} = q_{wall} \]  

(15)

where \( u_r \) and \( u_z \) are the axial and radial components of velocity, \( T_{in} \) is the inlet temperature, \( q_{rad} \) is the radiative flux profile on the cavity wall, and \( q_{wall} \) is the heat flux at the outer wall of the reaction zone. The baseline values for each boundary condition are listed in Table 3 at the beginning of section 6.

### 4. THERMOPHYSICAL PROPERTIES

The physical properties of CO\(_2\) and CaCO\(_3\) are obtained from the ANSYS Fluent materials database [26]. The complex refractive index of CaCO\(_3\) is obtained from the literature [27]. The physical properties of the SiC cavity walls are taken from CoorsTek data for reaction bonded silicon carbide and are assumed not to vary with temperature [28]. The emissivity, \( \varepsilon \), of SiC comes from Toloukian [29]. Specific values of thermophysical properties are shown in Table 2.
Table 2 Thermophysical properties

<table>
<thead>
<tr>
<th>Property</th>
<th>Material</th>
<th>CO₂</th>
<th>SiC</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \rho ) (kg m(^{-3}))</td>
<td>2800</td>
<td>1.78</td>
<td>3100</td>
</tr>
<tr>
<td>( k ) (W m(^{-1}) K(^{-1}))</td>
<td>2.25</td>
<td>0.0145</td>
<td>125(^1)</td>
</tr>
<tr>
<td>( c_p ) (J kg(^{-1}) K(^{-1}))</td>
<td>856</td>
<td>841.38</td>
<td>800</td>
</tr>
<tr>
<td>( \varepsilon )</td>
<td>–</td>
<td>–</td>
<td>0.9</td>
</tr>
<tr>
<td>( m )</td>
<td>1.55[1 + 0.004 \exp \left( -10 \mu \text{m}^2 \lambda^2 \right) ] + 0.1 \exp \left[ 0.2 \mu \text{m}^2 (\lambda^2 - 9 \mu \text{m}^2) \right] ]</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

\[^1\text{At 20°C.}\]

5. NUMERICAL SOLUTION

5.1. Mass, Momentum, and Energy Conservation Equations

The finite volume technique as implemented in the computational fluid dynamics (CFD) software ANSYS Fluent is used to solve the conservation equations. The continuity equation is solved using the projection method and the SIMPLE segregated pressure–velocity coupling algorithm. The momentum and energy equations are solved with a second-order upwind scheme. Gradients are evaluated using a least squares cell-based discretization scheme. Mesh independence is checked by increasing the number of CFD cells by 80%. This changes the solution by less than 5% for all parameters of interest.

5.2. Radiative Exchange in the Receiver Cavity

The radiation in the receiver cavity is modeled using the collision based Monte Carlo (MC) ray-tracing method. The in-house developed MC subroutine calculates the net heat flux at the inner cavity wall due to the solar source and reradiation inside the cavity [30]. The power of each ray and the numbers of rays launched at the aperture and the emitting cavity wall elements are calculated using:

\[
\dot{Q}_{\text{ray}} = \frac{\dot{Q}_{\text{solar}} + \varepsilon \sigma \sum_i A_i T_i^4}{N_{\text{rays}}} \quad N_{\text{solar}} = \frac{\dot{Q}_{\text{solar}}}{\dot{Q}_{\text{ray}}} \quad N_i = \frac{\varepsilon \sigma A_i T_i^4}{\dot{Q}_{\text{ray}}} \quad (16)
\]

where \( A_i \) is the emitting surface element area and \( N_{\text{rays}} \) is the total number of rays. The location and direction of a ray at the aperture assuming uniform emission are calculated as:
\[ \mathbf{\hat{r}} = [r\cos\phi, r\sin\phi, z_{\text{bottom}}] \], \( r = r_{\text{ap}}\sqrt{R} \), \( \phi = 2\pi R \) \hspace{1cm} (17)

The direction of the ray launched at the aperture is obtained from:

\[ \mathbf{\hat{u}} = [\sin\theta\cos\psi, \sin\theta\sin\psi, \cos\theta] \], \( \theta = \sin^{-1}\left(\frac{\sin\frac{\pi}{4}\sqrt{R}}{4}\right) \), \( \psi = 2\pi R \) \hspace{1cm} (18)

The location of ray emission by the cavity wall elements is obtained from

- for top and bottom surfaces

\[ \mathbf{\hat{r}} = \left[ r\cos\phi, r\sin\phi, \{H \text{ or } z_{\text{bottom}}\} \right] \], \( r = \sqrt{R\left(r_{i}^{2} - r_{i-1}^{2}\right)} + r_{i-1}^{2} \), \( r_{i} = \frac{r_{i+1}}{n_{r}} \), \( \phi = \mathcal{R}(\theta_{j} - \theta_{j-1}) + \theta_{j-1} \), \( \theta_{i} = \frac{2\pi j}{n_{\theta}} - \pi \) \hspace{1cm} (19)

- for cylindrical surface

\[ \mathbf{\hat{r}} = \left[ r_{\text{cav}}\cos(\phi), r_{\text{cav}}\sin(\phi), z \right] \], \( z = \mathcal{R}\left(z_{k} - z_{k-1}\right) + z_{k-1} \), \( z_{k} = \frac{Hk}{n_{z}} + z_{\text{bottom}} \), \( \phi = \mathcal{R}(\theta_{j} - \theta_{j-1}) + \theta_{j-1} \) \hspace{1cm} (20)

The direction of a ray emitted from the cavity walls is obtained by assuming grey and diffuse surfaces:

\[ \mathbf{\hat{u}} = \mathbf{\hat{n}} + \mathbf{\hat{t}}_{1}, \mathbf{\hat{n}} = \mathbf{\hat{n}}\cos(\theta), \mathbf{\hat{t}}_{1} = \sin(\theta)\left[\mathbf{\hat{t}}_{i}\cos(\psi) + \mathbf{\hat{t}}_{2}\sin(\psi)\right], \theta = \sin^{-1}(\sqrt{R}) \), \( \psi = 2\pi R \) \hspace{1cm} (21)

where \( \mathbf{\hat{n}}, \mathbf{\hat{t}}_{1}, \) and \( \mathbf{\hat{t}}_{2} \) are the normal and tangential vectors of the surface at the emission point. With the ray origin and direction defined, the rays are traced to the nearest intersection point with the aperture or the cavity walls. At the intersection point, a random number is generated. If it is less than the absorptivity of the surface, the ray is absorbed, the location of absorption is recorded, and a new ray is launched. Otherwise the ray is reflected and the direction of a diffusely reflected ray is determined using Eq. (21).

Ray tracing continues until the ray is absorbed or exits the reactor through the aperture. Once all rays from the aperture and cavity walls have been traced, the net heat flux to each surface is given by:

\[ q_{\text{rad,abs,em,ray}} = \frac{(N_{\text{abs,em}} - N_{\text{em}})\hat{Q}_{\text{ray}}}{A_{i}} \] \hspace{1cm} (22)

The MC simulation is executed on a structured, axisymmetric mesh with 10 radial, 16 angular, and 25 axial elements. The cavity wall temperature is passed from Fluent to the MC subroutine via a user defined function. Since the numerical mesh used in the MC simulations is coarser than the mesh used for the CFD computations, temperatures averaged over multiple CFD cells contained in MC cells are used in the MC simulations.

The net heat flux data for the top, bottom, and cylinder surfaces are passed back to Fluent by the user defined function for use as boundary conditions on the cavity wall.

6. RESULTS

The baseline simulation parameters used in this study are shown in Table 3. These parameters are used unless otherwise specified.
Table 3 Baseline simulation parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total number of rays</td>
<td>$N_{\text{total}}$</td>
<td>10,000,000</td>
</tr>
<tr>
<td>Inlet temperature (K)</td>
<td>$T_{\text{in}}$</td>
<td>300</td>
</tr>
<tr>
<td>Inlet velocity (m s$^{-1}$)</td>
<td>$u_{z,\text{in}}$</td>
<td>0.01</td>
</tr>
<tr>
<td>Outer wall heat flux</td>
<td>$q_{\text{wall}}$</td>
<td>0</td>
</tr>
</tbody>
</table>

Figure 3 shows the pressure drop through the reaction zone, which is related to the amount of pumping work needed to push the flowing gases through the packed bed. The pressure varies in the $z$ direction only. As the cavity diameter increases, the annular area of the reaction zone decreases and the pressure drop through the reaction zone increases. As the cavity length-to-diameter ratio increases, the reaction zone becomes longer and the pressure drop through the reaction zone also increases. To reduce the energy requirements for the process, the pressure drop across the bed should be minimized. A cavity with a smaller diameter and smaller length-to-diameter ratio minimizes the pressure drop.

The area-averaged axial temperature increase from the inlet to the outlet of the reaction zone is plotted in Fig. 4. Area-averaged temperature is defined as

$$T_{\text{av}} = \frac{1}{A} \sum_{i=1}^{n} T_i A_i$$

where $A_i$ is the area of a cell surface facet. The effect of the cavity diameter on the temperature drop does not have a clear upward or downward trend. The difference between different cavity diameters is less than 100 K and the difference between length-to-diameter ratios is between 38 and 52 K. A shorter cavity reduces the temperature difference slightly.

Figure 5 shows the area-averaged radial temperature drop across the reaction zone. The temperature drop is greatest for the smallest cavity diameter and decreases as the diameter is increased. This is due to the fixed reaction zone volume: as the cavity diameter and length-to-diameter ratio increase, the reaction zone becomes radially thinner. The higher length-to-diameter ratio lowers the radial temperature drop across the reaction zone by about 5–10 K, but the cavity diameter contributes a much larger reduction in temperature gradients. This temperature drop should be minimized in order to achieve an even reaction rate throughout the reaction zone.

![Fig. 3 Pressure drop through reaction zone](image)

![Fig. 4 Area-averaged axial temperature increase through reaction zone](image)
The area-averaged radial temperature drop across the cavity wall is displayed in Fig. 6. In all cases the temperature drop is less than 1 K due to the high thermal conductivity of silicon carbide. However, increasing the cavity diameter and length-to-diameter ratio while holding the reaction zone volume constant further reduces the temperature drop. This temperature drop should be minimized in order to reduce thermal stresses in the cavity wall during reactor operation.

Figure 7 shows the total heat transferred to the reaction zone at steady state. Ideally, the reaction zone would receive enough energy such that the reaction rate would not become heat transfer limited, so high rates of heat transfer are desired. The heat rate is highest for smaller cavity diameters and length-to-diameter ratios when the reaction zone volume and solar concentration ratio are fixed. This is due to the apparent emissivity of the cavity increasing as the cavity diameter and length-to-diameter ratio increase. Since the average cavity temperature between cases varies by less than 100 K and the aperture diameter remains the same, an increase in the effective emissivity results in an increase in radiative losses through the aperture and thus reduced heat transfer to the reaction zone.

Since reaction (1b) becomes thermodynamically favorable above 1150 K at atmospheric pressure, it is important that as much of the reaction zone as possible exceeds this temperature to avoid areas of unreacted particles. In all cases, the reaction zone reaches 1150 K within 5 mm of the inlet. This means that between 95% and 98.5% of the total reaction zone volume is usable for the calcination reaction, with the larger values occurring in the longer reaction zones.
7. CONCLUSIONS

A 1 kWth dual-cavity solar thermochemical reactor concept is proposed to capture carbon dioxide via the calcium oxide based calcination–carbonation cycle. The Monte Carlo ray-tracing method is employed to solve for radiative exchange in the inner cavity, coupled with a computational fluid dynamics analysis to solve the mass, momentum, and energy equations in the concentric reaction zone that is modeled as a gas-saturated porous medium consisting of optically large semitransparent particles.

The cavity diameter is varied from 6 to 10 cm and the length-to-diameter ratio is varied from 2 to 3 to study their effects on the pressure drop, temperature distribution, and heat transfer in the reactor. Increasing the cavity diameter and length-to-diameter ratio decreases the radial temperature gradients across the cavity wall and reaction zone, reducing thermal stresses in the cavity wall and helping ensure uniform reaction rates. However, increasing the cavity diameter also results in increased pressure drop through the reaction zone and reduced heat transfer to the reaction zone. Changing the cavity diameter has a greater effect than changing the length-to-diameter ratio. Because of these competing effects, a moderate cavity diameter of 8 cm is chosen for this reactor.

ACKNOWLEDGEMENT

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NOMENCLATURE

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<td>$C_f$</td>
<td>Forchheimer coefficient, m$^{-1}$</td>
</tr>
<tr>
<td>CFD</td>
<td>computational fluid dynamics</td>
</tr>
<tr>
<td>$f_{vs}$</td>
<td>solid volume fraction</td>
</tr>
<tr>
<td>$H$</td>
<td>cavity height, m</td>
</tr>
<tr>
<td>$\hat{i}$</td>
<td>unit vector in r direction</td>
</tr>
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REFERENCES


