2011 International Conference on Environmental Science and Engineering
(ICESE 2011)

Study on the Radiation Distribution in A Fluidized Tubular Reactor for Heterogeneous Photocatalytic Hydrogen Production

Zhao Liang*, Fan Liangliang, Jing Dengwei, Liu Wenxu, Cao Fei, Zhu Xule, Yang Junyin, Zhang Hailong

State Key Laboratory of Multiphase Flow in Power Engineering, Xi’an Jiaotong University Xi’an, P.R.China
lzhao@mail.xjtu.edu.cn

Abstract
Radiation distribution study is of importance for the design and the optimization of fluidized photocatalytic tubular reactor which has been developed for photocatalytic hydrogen production under direct solar light. In the present study, the radiation distribution in such reactor was successfully simulated by adopting Monte Carlo method and the six-flux radiation absorption-scattering models. Both the incident angles of light around the reactor and the concentration distribution of photocatalyst were taken into account. Our analysis indicates that the angles and the intensity of the incident rays significantly affect the radiation distribution. Moreover, it was also found that the equilibrium radiation distribution has a close relationship with the density distribution of the photocatalysts. The simulated results are expected to be helpful for obtaining the optimal operating parameters for solar photocatalytic hydrogen production.

© 2011 Published by Elsevier Ltd. Selection and/or peer-review under responsibility of [name organizer]
Open access under CC BY-NC-ND license.
Keywords: Radiation distribution, Fluidized tubular reactor, Nanometer catalyst, Compound parabolic concentrators

1. Introduction

Conventional energy resources such as coal and petroleum products have been depleted to a great extent. It is therefore necessary to produce an alternative fuel that should in principle be pollution free, storable and economical. Hydrogen satisfies the first two conditions and to fulfill the third requirement. Photocatalytic hydrogen production from water using solar energy has been identified as a promising technology for the final realization of renewable mass hydrogen production. However; many problems must be addressed before this technology become economically viable. One issue is the development of
efficient visible light driven photocatalyst which has undergone a rapid progress over the past years [1-2]. The other key issue concerns the efficient utilization of the solar energy itself. The design of the photocatalytic reactor is closely related to this aspect.

Many different photocatalytic reactors have been developed, such as parabolic-trough concentrators (PTC) [3], thin-film-fixed-bed reactor (TFFBR) [4-5], double skin sheet reactor (DSSR) [6] and compound parabolic concentrators (CPC) [7]. CPC needs no light tracking system and is a good option for solar photochemical applications [8-13]. In despite of so many types of solar reactors designed for solar detoxification purposes, few attempts have been made for photocatalytic hydrogen production. Addressing both the similarity and dissimilarity for these two processes and by fully considering the special requirements for the latter reaction, we have designed a Compound Parabolic Concentrator (CPC) based photocatalytic hydrogen production solar reactor for the first time in our previous work, as shown in Fig 1[14].

![Figure 1 The system of photocatalytic hydrogen production](image)

To improve and optimize solar photocatalytic hydrogen production system, the optimal utilization of the incident solar light is considered to be very important. Radiation field distribution in the photocatalytic reactor is therefore indispensable for evaluating the designed reactor. While it is accepted that direct and accurate measurement of the radiation field is very difficult, numerical model may provide another effective tool for this target. Much work on radiation field models has been developed. Alexiadis et al. studied the modeling of a photocatalytic reactor with a fixed bed of supported catalyst and PTR reactor [15-17]. However, work on the radiation distribution in a fluidized tubular reactor using CPC has not been attempted. In this study, the radiation distribution in the fluidized tubular reactor was successfully simulated. Besides, we used the result of the model to estimate the efficiency of hydrogen production and obtained the result which was comparable to the experimental results.

![Figure 2 Six-flux radiation absorption-scattering model](image)
2. Modeling of the Fluidized Tubular Reactor

2.1 Monte Carlo Simulation and Six-flux Radiation Absorption-scattering Model

In the work, we used the six-flux radiation absorption-scattering model which could be described as the scattered photons follow the route of the six directions of the Cartesian coordinate. The six-flux radiation absorption-scattering model is shown in Fig.2. A volume element ($\Delta V$) in which the photons traveled in the axial direction, as schematically illustrated in Fig 3, was proposed. The catalysts are supposed not overlapping. The sectional area of radiation can be described as follows:

$$s = \frac{1}{4} \pi d^2 \times n \times \Delta V$$  \hspace{1cm} (1)

where $d$ is the averaged diameter of the catalyst, and $n$ the number of the catalyst in $\Delta V$.

$\xi$ indicating the probability of photons absorbed by catalyst can be expressed as the following equation:

$$\xi = \frac{s}{\Delta S} = \frac{1}{4} \pi d^2 \times n \times l$$  \hspace{1cm} (2)

Supposed that the concentration of the catalyst in the reactor is $p_0$, the mass density of the catalyst is $p$ and the catalyst is simplified as a miniature ball. Then we can compute the average volume that each catalyst takes up in the reactor as follows:

$$V_0 = \frac{1}{n} \frac{1}{6} \frac{\pi d^3}{p_0}$$  \hspace{1cm} (3)

$l$ can be obtained by the following equation:

$$l = \frac{s_0}{\frac{1}{4} \pi d^2} = 4 \left(\frac{D \pi}{6 \rho_0}\right)^\frac{2}{3}$$  \hspace{1cm} (4)

$\xi$ can be obtained by another equation as follows:

$$\xi = \frac{3 \rho_0 l}{2 \rho d^2} = \pi \left(\frac{6 \rho_0}{\rho}\right)^\frac{1}{3}$$  \hspace{1cm} (5)

2.2 CPC Model and the Concentration Distribution of Catalyst in the Reactor

We have studied on the intensity distribution of the incident light around the outer surface of CPC. The intensity distribution of the incident light around the CPC we employed in this work is shown in Fig.4. The concentration distribution of the catalyst in the fluidized tubular reactor on different conditions was studied in our previous work [18]. Based on that work, we take into account the uneven distribution of the
catalyst in the tube at different flow rate, and simulate the radiation field under different conditions.

![Figure 4 The intensity distribution of the incident light around the CPC](image)

### 2.3 Simplification and Assumptions for the Model

1) The external surface of the reactor does not reflect the incident light.
2) When the photon runs into the catalyst, the properties of the ray and the catalyst do not change.
3) The catalyst particles do not overlap or agglomerate.
4) There are 360 positions where the incident ray propagates into the reactor.
5) There are 100000 rays used in the simulation.

### 3. Results and Discussion

#### 3.1 Radiation distribution in simplified cases

The radiation distribution for the cross section of the tube obtained in simulation is shown in Fig.5. The radiation intensity along the vertical center line the cross section is shown in Fig.6.

![Figure 5 The radiation distribution in model 1](image)

![Figure 6 The radiation distribution in the vertical middle axle of the reactor](image)
As shown by Fig. 6, the radiation distribution in the reactor has a close relationship with the concentration of the catalyst. The radiation intensity is strong in the annular region near the internal surface of the reactor, and decreases with the increase of the distance from the internal surface of the reactor. There is an intensity peak in the center of the reactor. As shown in Fig. 6, this is reasonable considering that all the incident lights are pointed to the focus of the annulus.

As a further step, four different catalyst concentration distributions were considered. The change of the peak’s value we obtained from Fig. 7 is shown in Fig. 8.

![Figure 6](image)

(a)                         (b)
![Figure 6](image)

(c)                         (d)

Figure 6 The different radiation distributions under different concentration distributions. (a) 1.5\(\text{m/s}\), (b) 2.0\(\text{m/s}\), (c) 2.5\(\text{m/s}\), (d) 3.0\(\text{m/s}\)

3.2 Radiation distribution considering the reflections of CPC

As a simulation on the practical case of our solar reactor, reflection of CPC and the different concentration distribution of catalyst were considered this time. The direction and the intensity of the incident ray around the reactor were also tracked. Considering the concentration of the catalyst to be 0.5\(\text{g/L}\) and 2.0\(\text{g/L}\), respectively, we obtained the effect of the different concentration but the same concentration gradient. The data is shown in Table 1.

As shown in Fig. 9, the concentration of the catalyst has an effect on the radiation distribution in the reactor, similar to the case without CPC. It is found that when the catalyst concentration is 0.5\(\text{g/L}\), the penetration of the photon flux become quite deeper. It also shows that regardless of the catalyst concentration and the flow rate, the solar intensities are much stronger at mainly three parts of the tube. That means, if we can load the photocatalysts especially at these parts of the reactor tube, the enhanced utilization efficiency of the catalyst and the improved hydrogen production are expected.

![Figure 7](image)

Figure 7 The change of the peaks’ value. 1 the peak value in model 1, 2 \(v=1.5\text{m/s}\), 3 \(v=2.0\text{m/s}\), 4 \(v=2.5\text{m/s}\), 5 \(v=3.0\text{m/s}\). peak 1 the left peak of radiation intensity of the vertical middle axle of the reactor, peak 2 the middle peak, peak 3 the right peak.
Figure 8 The radiation distribution in the reactor when the concentration of catalyst is (a) 0.5g/L; (b) 2.0g/L

Table 1 Photo utilization conditions

<table>
<thead>
<tr>
<th>Subject</th>
<th>Number</th>
</tr>
</thead>
<tbody>
<tr>
<td>The concentration of the catalyst</td>
<td>0.5</td>
</tr>
<tr>
<td>The number of photons absorbed by the catalyst</td>
<td>73903</td>
</tr>
<tr>
<td>The number of photons that escape from the reactor</td>
<td>26097</td>
</tr>
<tr>
<td>The velocity in the model</td>
<td>1.5</td>
</tr>
</tbody>
</table>

4. Hydrogen Production Efficiency Prediction and Comparison to the Experimental Results

The catalyst we used in hydrogen production efficiency prediction can only use the UV and some of the visible light of the sun. It means the proportion of the sunlight that can be used by the catalyst is between 5% and 48% because the UV takes up 5% of the sunlight and the visible light takes up 43%. Firstly, we supposed that 5% of the sunlight would be used by the catalyst, the utilization rate of the photons is 79.83% which was obtained in model 4, the solar irradiance (I) is 1kW/m², the area (S) of the CPC is 1.6m², and the efficiency of light energy conversion (ƞc) is 6.1% obtained from the experiment. The effective energy of sunlight (Es) used by the hydrogen production system was calculated using:

\[ E_s = I \times S \times 5\% \times 79.83\% = 63.9W \]  \hspace{1cm} (6)

The hydrogen production rate (Rp) was calculated using the following formula:

\[ R_p = \frac{E_s \eta_c}{\Delta G^0_p}. \]  \hspace{1cm} (7)

If we change the proportion of the sunlight that can be used by the catalyst to be 48%, we can get the hydrogen production of 12.67L/h in our experimental result, the measured hydrogen production rate is 1.88L/h. The two results are comparable. In different models of our work, the hydrogen production efficiencies are different. We figured out the different hydrogen production efficiency on condition that the concentration is 1g/L and the proportion of the sunlight that can be used by the catalyst is 48%. The results are shown in Table 2.

In Table 2, it is found that the hydrogen production efficiency has a close relationship with CPC and concentration distribution of catalyst. In contrast to the previous simulation, the CPC and the concentration distribution of catalyst make the result more comparable to the experimental result.
Table 2 The hydrogen production efficiency for different conditions

<table>
<thead>
<tr>
<th>Modeling conditions</th>
<th>Utilization rate of photons (%)</th>
<th>Hydrogen production efficiency (g/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model 1 (simple)</td>
<td>84.7</td>
<td>13.44</td>
</tr>
<tr>
<td>Model 2 without CPC (v=1.5m/s)</td>
<td>84.64</td>
<td>13.43</td>
</tr>
<tr>
<td>Model 2 without CPC (v=2.0m/s)</td>
<td>84.68</td>
<td>13.44</td>
</tr>
<tr>
<td>Model 2 without CPC (v=2.5m/s)</td>
<td>84.75</td>
<td>13.45</td>
</tr>
<tr>
<td>Model 2 without CPC (v=3.0m/s)</td>
<td>84.68</td>
<td>13.44</td>
</tr>
<tr>
<td>Model 3 with CPC</td>
<td>80.02</td>
<td>12.70</td>
</tr>
<tr>
<td>Model 4 with CPC (v=1.5m/s)</td>
<td>79.83</td>
<td>12.67</td>
</tr>
<tr>
<td>Model 4 with CPC (v=2.0m/s)</td>
<td>79.86</td>
<td>12.68</td>
</tr>
<tr>
<td>Model 4 with CPC (v=2.5m/s)</td>
<td>80.17</td>
<td>12.72</td>
</tr>
<tr>
<td>Model 4 with CPC (v=3.0m/s)</td>
<td>79.89</td>
<td>12.67</td>
</tr>
</tbody>
</table>

5. Conclusions

In the work, we simulated different models to obtain the effects of CPC, the concentration of the catalyst and the concentration distribution of catalyst on the radiation distribution in the reactor. The results may be of great importance in designing a more efficient reactor and obtaining the optimal flow velocity for photocatalytic hydrogen production. In addition, the radiation distribution in a fluidized tubular reactor could be used to directly estimate the efficiency of hydrogen production. Based on the numerical results, the following conclusions can be made:

1) The CPC affects the radiation distribution greatly. The radiation intensity in the left lower, the right lower and the bottom regions of the reactor is strong.
2) The concentration distribution has a close relationship with the radiation distribution in the reactor, but the effect of the concentration distribution is less than that of the CPC.
3) There is an optimal velocity for the system of hydrogen production. The concentration distribution of the catalyst is the best for the utilization rate in the optimal velocity. In our work, the optimal velocity is 2.5m/s.
4) There is an optimal concentration of catalyst for the hydrogen production. In our work, while the concentration of catalyst is 1g/L, the result is the best.

6. Acknowledgments

The authors gratefully acknowledge the financial support of the National Natural Science Foundation of China (Grant No. 50821064), the National Basic Research Program of China (Grant No. 2009CB220000) and the National 863 Project of China (No. 2009AA05Z117).

References


