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Integrated simulation of turbulent convection, radiation and conduction during a selenization process for large-scale CIGS thin films

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ABSTRACT

A numerical methodology for simulations of multi-mode heat transfer during a selenization process for CIGS (Copper Indium Gallium Selenide) solar cell films has been developed. Turbulent fluid dynamics and convective heat transfer are simulated using a finite-volume large-eddy simulation (LES) technique while thermal conduction and radiation are predicted using finite-element methods. The computational methodology is validated for three heat transfer modes. Using the technique, a numerical study of heat transfer during the selenization process for deposition of a CIGS layer is performed to analyze the thermo-fluid phenomena occurring during the process. The present method is found to well predict temperature distribution on substrates as a function of both space and time. It is also analyzed that how turbulent fluid motions alter temperature distributions on the substrate during the selenization process.

1. Introduction

CIGS solar cells are composed of a light absorption layer, of which chemical compounds are Cu, In, Ga, and Se. A CIGS layer has a high photon absorption coefficient and efficiently absorb sunlight even with a highly thin film thickness (2–3 μm), which makes the CIGS solar cells have superior advantages to silicon solar cells in terms of the price. Since the efficiency of a solar cell is determined by the uniformity of the light absorption layer, a deposition process of the layer is regarded as the most important step during the solar cell manufacturing process.

Much research has been conducted to improve the light conversion efficiency of the light absorption layer. Repins et al. [1] used a three-stage co-evaporation process for the deposition of a CIGS layer. In the first stage, an $(In_{0.7}, Ga_{0.3})_2Se_3$ layer is grown by co-evaporation of In, Ga, and Se. In the second stage, a CIGS layer is formed by evaporating Cu and Se on the $(In,Ga)_2Se_3$ layer. In the third stage, In, Ga, and Se are evaporated on the CIGS layer to change into a Cu-poor film [2]. Using the three-stage co-evaporation process, Repins et al. [1] were able to produce a CIGS solar cell with efficiency of 19.9% and fill factor of 81.2%. The fill factor indicates the solar cell performance and is defined as a ratio of the maximum solar cell power to the product of the open circuit voltage and the short circuit current. Later, Jackson et al. [3] further improved the CIGS solar cell efficiency to 21.7%. These studies, however, were carried out for lab-scale cells with sizes of less than 1 cm^2. For small scale cells, accurate control of the composition and temperature of involving chemical species are rather easily achievable.

In spite of the high efficiency in the lab-scale research, efficiency enhancement has been rather slow for practical scale CIGS solar cells. Powalla and Dimmler [4] produced a CIGS solar cell with the size of 30 cm × 30 cm with conversion efficiency of 12.7% using an in-line process. Although the in-line process is considered as a good solution for mass production, the conversion efficiency of solar cells produced by the in-line process is found still rather low. Furthermore, it is known that supplement of thermal energy and control of chemical composition on substrates are difficult using the in-line process [5].

A two-step process is considered suitable for production of practical size CIGS solar cells [6,7]. The process consists of two-step processes: a sputtering step and a selenization step. In the sputtering step, Cu, In, and Ga are ejected from sources of solid materials. In the selenization step, hydrogen selenium gas which is highly reactive and toxic reacts with metal precursors and finally forms a CIGS thin film. After the selenization process, the surface of a substrate often shows spatially non-uniform deposition of the chemical species indicated by the darkness of the stain. If severe non-uniform stains are formed, the conversion efficiency of the substrate is also degraded. Therefore if causes of the non-uniform stains are identified, the conversion efficiency of CIGS solar cells can be further improved. Due to the toxicity of hydrogen selenium,

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Nomenclature

Roman symbols

Bi	Biot number, hL/k_{solid}
C_p	heat capacity
F	view factor
g	gravitational acceleration
Gr_x	Grashof number, $g\beta(T_{wall}-T_\infty)L^3/\nu^2$
h	convective heat transfer coefficient
k	thermal conductivity
L	characteristic length
Nu	Nusselt number, hL/k_{fluid}
Pr	Prandtl number, ν/α
q	heat flux
Ra_x	Rayleigh number, $g\beta(T_{wall}-T_\infty)x^3/\nu\alpha$
T	temperature
U	x-directional velocity component
x, y, z	Cartesian coordinates

Greek symbols

α	thermal diffusivity
α_{rad}	radiative absorptivity
β	thermal expansion coefficient
ϵ	radiative emissivity
η	dimensionless distance, $y(\nu x/U_\infty)^{-1/2}$
ν	kinematic viscosity
ρ	fluid density
σ	Stefan-Boltzmann constant
τ	selenization process time
θ	normalized temperature, $(T-T_\infty)/(T_h-T_\infty)$
ζ	dimensionless transverse coordinate, $-y(\partial\theta/\partial y)_{y=0}$

Subscripts

heater	surface of the heater
sub	surface of the substrate

however, it is extremely difficult to observe the physical phenomena occurring during the process and to analyze factors affecting the conversion efficiency of solar cells. Therefore, an accurate numerical method is needed to predict and understand physical phenomena occurring during the selenization process.

In the present study, a numerical methodology for studying the selenization process is developed. Unsteady turbulent fluid dynamics and convective heat transfer are predicted using a large-eddy simulation (LES) technique while thermal conduction and radiation are predicted using finite-element methods (FEM). First, the present method is validated using natural convection in a vertical flat plate and natural convection with surface radiation in a side-vented open cavity. The present methodology is applied to a simulation of heat transfer during a large-scale selenization process. Averaged temperature of substrates predicted by the present method is compared to experimental results.

The computational methodology of the present study is introduced in Section 2. In Section 3, the present numerical method is validated for test problems involving multiple heat transfer modes. Details of the computational configuration and setup for simulations of a large-scale selenization process, and simulation results compared to the experimental measurements are presented in Sections 4 and 5, respectively, followed by concluding remarks in Section 6.

2. Computational methodology

Governing equations for fluid motions are incompressible Navier-Stokes equations as follows:

$$\frac{\partial u_i}{\partial x_i}=0, \tag{1}$$

$$\frac{\partial u_i}{\partial t}+\frac{\partial u_i u_j}{\partial x_j}=-\frac{1}{\rho}\frac{\partial p}{\partial x_i}+\nu\frac{\partial^2 u_i}{\partial x_j\partial x_j}+g_i\beta(T-T_\infty), \tag{2}$$

$$\frac{\partial T}{\partial t}+u_j\frac{\partial T}{\partial x_j}=\alpha\frac{\partial^2 T}{\partial x_j\partial x_j}, \tag{3}$$

where g_i is the gravitational acceleration, ρ is the fluid density, ν is the kinematic viscosity, β is the thermal expansion coefficient, and α is the thermal diffusivity. The third term on the right-hand side of Eq. (2) accounts for the buoyancy due to the temperature difference of the fluid. A finite-volume method is used to discretize the governing equations. Governing equations are discretized with second-order accurate schemes in time and space. Numerical algorithms and solution methods are described in detail in Ref. [8]. A dynamic global-coefficient subgrid-scale model is used for subgrid scale turbulence closure in LES [9].

Finite-element methods are used to calculate conductive and radiative heat transfer in a thermal reactor which is geometrically complex. The structure domain is divided into smaller components where the heat transfer equation is expressed in an integral form. Each element which has a hexahedral shape contains 20 nodes that are located at the vertices and in the middle of edges. These elements ensure better accuracy. A backward Euler scheme is used for the time integration. The governing equation for the unsteady heat conduction is as follows:

$$-k\frac{\partial^2 T}{\partial x_j\partial x_j}+\rho C_p\frac{\partial T}{\partial t}=q_v, \tag{4}$$

where k is the thermal conductivity, C_p is the heat capacity, and q_v is the heat flux. Thermal radiation is considered to be cavity radiation because operating gases are optically thin. In order to consider thermal radiation inside the reactor, the view factor is used. The view factor F_{ij} refers to the ratio of radiant power leaving surface A_i, to absorbing surface A_j, as follows [10]:

$$F_{ij}=\frac{1}{A_i}\int_{A_i}\int_{A_j}\left(\frac{\cos\psi_i\cos\psi_j}{\pi R^2}\right)dA_i dA_j, \tag{5}$$

where R is the total radiation leaving the surface A_i, ψ_i, and ψ_j are angles. Fig. 1 shows the configuration for the view factor calculation. The view factor calculation requires a large amount of computational operations. Since the shape of the thermal reactor does not change, the view factor is calculated once at the beginning of the simulation. The radiative heat flux equation is as follows:

$$q_{rad}=\epsilon\sigma T^4-\alpha_{rad}\sigma T_\infty^4, \tag{6}$$

where ϵ is the emissivity, α_{rad} is the absorptivity of thermal radiation, and σ is the Stefan-Boltzmann constant.

Fig. 2 shows heat exchanges between the heater and the surround. It is assumed that the surface is gray, independent of wavelength, diffuse, independent of direction. It is also assumed that