Numerical study of a chemical method applied to instantaneous heat removal under high heat flux

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Abstract In this study, a method of chemical cooling is put forward, that is, C–CO₂ endothermic reaction is applied to instantaneous heat removal under high heat flux. A method in which theoretical research is in combination with numerical simulation is used to study C–CO₂ endothermic reaction. In comparison with the theoretically computational results, numerical code is validated. A high heat flux of 500 W/cm² is applied to the research of the heat dissipation characteristics of C–CO₂ endothermic reaction. The theoretical calculation results show that, under a certain temperature and pressure condition, the C–CO₂ chemical endothermic reaction could remove heat from the system promptly; the product CO could be used as a supplementary medium of power source for cycling. Compared with water phase change, the C–CO₂ endothermic reaction appears to have stronger heat removal ability. “Species Transport” module in FLUENT was adopted to simulate the reaction. Under the same temperature and pressure condition, the numerical simulation results are found to be well congruous with theoretical results. The C–CO₂ endothermic reaction could make a high temperature in the reaction system due to a high heat

flux reduce to a low temperature (below zero) promptly. The heat removal and reaction time are in consistence with theoretical calculation.

List of symbols

A	Pre-exponential factor (1/s)
c	Reactant concentration (mol/m ³)
C_p	Heat capacity (J/kg K)
$D_{I,m}$	Mass diffusivity of gas I in gas mixture (m ² /s)
E_a	Activation energy (kJ/mol)
m	Mass (kg)
n	Reaction order (–)
P	Pressure (Pa)
q	Volumetric heat source (W/m ³)
Q	Heat (KJ)
R	Gas constant (J/mol K)
S_m	Mass source term
t	Time
T	Temperature (K)
u, v	Velocities in axial and radial directions, respectively (m/s)
U	Velocity vector in the x-direction and y-direction, u and v
x	Coordinate in axial direction (m)
y	Radial coordinate (m)
Y	Mass fraction (%)

Greek letters

κ	Reaction rate constant
θ	Surface fraction
β	Absorption capacity
γ	Reaction rate
ρ	Density (kg/m ³)
μ	Dynamic viscosity (kg/m*s)
λ	Thermal conductivity (W/m* °C)

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1 Introduction

With the development of new high-techs, occasions due to high and even ultra high heat flux appear. High heat flux results in high temperatures, which need higher requirements for the property of material and operating conditions and produce a negative effect on the service life-span of material. So heat removal problem caused by dense situation of high heat flux becomes increasingly serious. For example, ablation on the electrode surface caused by high heat flux, not only results in degradation of the overall performance of equipment, but also produces a negative effect on the service lifespan of equipment [1, 2]; in order to improve the thermal efficiency and power output, advanced gas turbine engines usually work under high temperatures of 1,300–1,500 °C, which cause the operating temperature of turbine blade to be much higher than that of metal materials allowable temperature, so reasonable cooling for blade becomes a problem which we have to be faced with [3]; heat flux density on the thermal deposit surface of high power semiconductor laser can reach as high as 400 W/cm², which is limited to a certain level of performance and further improvement by heat removal problem [4]; temperature of hypersonic aircraft (Mach number is 8 or above) head can reach 2,300 °C, and combustion indoor temperature can reach above 3,000 °C [5].

Some mature forced cooling methods at present are based on physical heat sink, including forced air cooling, water phase change, pool boiling, spray cooling, micro-channel cooling and micro-jet array cooling et al.

Forced air cooling [6] is a physical cooling method which uses the fan to blast or ventilate to improve air flow rate to achieve the purpose of cooling. It is widely used in high-power electronic device field, because its heat transfer capacity is several times more than that of natural air cooling, it is, however, rarely applied to LED thermal design but only applied to electronic devices whose outer surface heat flux density does not exceed 10 W/cm² due to its complex system, heavy noise, low reliability, expensive maintenance costs and extra power dissipation.

Pool boiling, also known as large container boiling, which is one of liquid boiling methods in industry, is to immerse the heating surface into liquid, and the liquid heats on the wall surface. Pool boiling is greatly valuable in the field of industrial application. In the past few decades, the scholars in different countries have done a lot of research by conducting many boiling heat transfer experiments and put forward various heat transfer mechanism models, but due to the limitations of experiments and assumptions, these models have their own defects [7].

Water phase change cooling, which is based on physical phase change, can only provide 67 kJ/mol of heat transfer

capacity; the most commonly used gravity heat pipe can only provide <30 W/cm² of heat transfer capacity.

In some extreme physical situations, such as super high temperature reaction kettle, super high temperature gas turbine engine, super mach number propeller, high power laser pump and so on, physical heat sink cooling methods [8] based on physical phase change of working medium are limited by the latent heat of vaporization ability and unable to meet the requirement of heat removal due to high heat flux density. Therefore, since the 1960s, the United States has into practice the study of chemical heat sink application as cooling method [9]. Many studies focused on how to make the chemical endothermic reaction applied to heat-dissipation problem [10–12].

The reaction of ethanol dehydration to ethylene is an endothermic reaction with good thermal effect. In the preparation of ethylene, by using the adiabatic tube reactor, heat could be removed from the reactor with the endothermic reaction, while coke formation is avoided.

Through endothermic reaction, heat could be removed from a low temperature heat source, and through exothermic reaction, heat could be upgraded to heat sink at a high temperature. Karaca et al. [13] studied the chemical heat pump system made up with methanol–formaldehyde–hydrogen, ethanol–acetaldehyde–hydrogen, propanol–acetone–hydrogen, butanol–butyraldehyde–hydrogen and so on at a low temperature. In the system, dehydrogenation is endothermic while hydrogenation is exothermic. On the basis of economic analysis, a large amount of waste heat is greatly needed to achieve the thermal efficiency.

A new chemical approach, which use C–CO₂ endothermic reaction applied to instantaneous heat removal in a high heat flux density situation, is put forward in this paper, which is rarely reported currently at home and abroad. Li et al. [14] explored the effect of CO₂ on hard coke degradation reaction, but they did not study the method of application of C–CO₂ endothermic reaction to instantaneous heat removal. In this paper, both theoretical calculation and numerical simulation methods are applied to the study of C–CO₂ endothermic chemical reaction. Both of the results showed that C–CO₂ endothermic chemical reaction could help realize instantaneous heat removal under high heat flux.

2 Theoretical calculation

The aim of this study is going to verify that instantaneous heat removal can be realized by C–CO₂ endothermic reaction. In the study, activated carbon particles and carbon dioxide gas are used as reactants. As the reaction proceeds, high energy renewable material-CO would be produced. CO could be used as supplementary medium of power

source, providing energy through combustion exothermic reaction, the product-CO₂ could be used again as a reactant of endothermic reaction. Thus a material cycle will form through above process. Because of the strong heat absorption capacity and the special relationship between reactants and products, this thinking will help realize instantaneous heat removal and material cycle.

Wood activated carbon is to be used in a subsequent experiment, whose specific surface area is more than 1,000 m²/g. Thus CO₂ can diffuse to internal of activated carbon particle fully for its porosity, which means activated carbon particles will react with CO₂ directly. Therefore, the influence of diffusion on reaction rate can be neglected.

C–CO₂ reaction can be simplified as follows [15]:

According to solid surface reaction model, two rate equations can be concluded:

$$\frac{dP_{\text{CO}}}{dt} = \kappa_1 P_{\text{CO}_2} (1 - \theta) + \kappa_2 \theta \quad (1)$$

$$\frac{d\theta}{dt} = \frac{1}{\beta} [\kappa_1 P_{\text{CO}_2} (1 - \theta) - \kappa_2 \theta] \quad (2)$$

In the equations, P_{CO} and P_{CO_2} refers to partial pressures of CO and CO₂, κ_1 and κ_2 are the reaction rate constant of reaction (A) and (B), θ is to refer to surface fraction occupied by surface oxide [CO]. β means absorption capacity.

At a high temperature, C–CO₂ reaction can be considered as first-order reaction, and starting from 800 °C, order of reaction will be close to 1. The higher the temperature is, the closer to the first-order reaction. A conclusion can be drawn from the above two equations. The higher the temperature is, the θ is closer to 0. Then reaction rate is: $\frac{dP_{\text{CO}}}{dt} = \kappa_2 P_{\text{CO}_2}$, which means reaction rate is proportional to P_{CO_2} ($n = 1$); the lower the temperature is, the θ is closer to 1. Then the reaction rate is: $\frac{dP_{\text{CO}}}{dt} = \kappa_2 \theta$, which means the reaction rate is independent of P_{CO_2} ($n = 0$).

C–CO₂ reaction is a strong endothermic one, its temperature produces a slight effect on activation energy. Table 1 shows the pre-exponential factor A and reaction activation energy E_a [16].

According to Arrhenius formula and chemical reaction rate equation,

Table 1 Reaction parameters

Reaction	Pre-exponential factor $A/1/\text{s}$	Activation energy $E_a/$ (kJ/mol)
$\text{CO}_2 + \text{C} = 2\text{CO}$	5.55×10^3	306.1

$$\kappa = A e^{-\frac{E_a}{RT}} \quad (3)$$

$$\gamma = \kappa \cdot c \quad (4)$$

Time for removing 500 W/cm² heat at different temperatures can be calculated. In the reaction, CO₂ concentration is used.

Table 2 and Fig. 1 show time for removing 500 W/cm² heat at different temperatures with C–CO₂ endothermic reaction.

From Table 2, it can be seen that, the higher a reaction temperature is, the less time it needs. Time for completing the endothermic reaction can reach the level of one percent second at a certain temperature (about 1,573 K).

To illustrate the strong endothermic capacity of C–CO₂ reaction, a comparison can be made between heat removal capacity of C–CO₂ endothermic reaction and water phase change under the same conditions (the same temperature, pressure and molar amount).

Air density is 1.225 kg/m³, heat capacity is 1.006 kJ/kg °C under constant pressure (from FLUENT database). Length of cylindrical reactor used in numerical simulation is 2 m, and its bottom radius is 0.225 m. If hot air is cooled from 1,500 to 300 K, heat to be removed is 470.35 kJ. Reaction equations of C–CO₂ reaction and water phase change are as follows:

Table 2 Relationship between reaction temperature and time

Temperature/ K	973	1,073	1,173	1,273	1,373	1,473	1,573
Time/s	23	6.37	2	0.78	0.37	0.17	0.09

Fig. 1 The graph of time variation with reaction temperatures

If Q is of positive value, the reaction is endothermic. To remove 470.35 kJ heat, CO_2 needed is 2.73 mol, while only 182.7 kJ heat can be removed through water phase change if the amount of molar water is the same, which means the temperature of hot air can only be cooled to 1,034 K. In comparison of heat absorption capacity of C– CO_2 reaction with water phase change, the former is much stronger than the latter. At the same time, the calorific value of CO, the product, is about 6 % higher, which is equivalent to transfer energy from one medium to the other. CO can be used as a high calorific value fuel for cycle, which provides necessary conditions for energy cycle.

3 Numerical simulation

3.1 Governing equations

An axial-symmetrical two-dimensional (x–y) computational domain is profiled as shown in Fig. 2, which includes two flow streams. The model has the following dimensions: Diameter of inlet-1 and inlet-2 are 0.01 and 0.45 m, respectively; thickness of the sheet is 0.005 m.

The following governing equations, Eqs. (5)–(10), for mass, momentum, energy and species transfer are universally applicable to the entire computational domain. However, zero velocities need to be assigned to the solid area in the numerical treatment.

Mass equation [17]:

$$\frac{\partial \rho}{\partial t} + \text{div}(\rho U) = 0 \quad (5)$$

Momentum equation [18]:

$$\frac{\partial(\rho u)}{\partial x} + \frac{1}{y} \frac{\partial(y \rho v)}{\partial y} = 0 \quad (6)$$

$$\begin{aligned} \frac{\partial(\rho u u)}{\partial x} + \frac{1}{y} \frac{\partial(y \rho v u)}{\partial y} = & -\frac{\partial P}{\partial x} + \frac{\partial}{\partial x} \left(\mu \frac{\partial v}{\partial x} \right) + \frac{1}{y} \frac{\partial}{\partial y} \left(y \mu \frac{\partial u}{\partial y} \right) \\ & + \frac{\partial}{\partial x} \left(\mu \frac{\partial u}{\partial x} \right) + \frac{1}{y} \frac{\partial}{\partial y} \left(y \mu \frac{\partial v}{\partial x} \right) \end{aligned} \quad (7)$$

$$\begin{aligned} \frac{\partial(\rho u v)}{\partial x} + \frac{1}{y} \frac{\partial(y \rho v v)}{\partial y} = & -\frac{\partial P}{\partial y} + \frac{\partial}{\partial x} \left(\mu \frac{\partial v}{\partial x} \right) + \frac{1}{y} \frac{\partial}{\partial y} \left(y \mu \frac{\partial u}{\partial y} \right) \\ & + \frac{\partial}{\partial x} \left(\mu \frac{\partial u}{\partial y} \right) + \frac{1}{y} \frac{\partial}{\partial y} \left(y \mu \frac{\partial v}{\partial y} \right) - \frac{2\mu v}{y^2} \end{aligned} \quad (8)$$

Energy equation [18]:

$$\begin{aligned} \frac{\partial(\rho C_p u T)}{\partial x} + \frac{1}{y} \frac{\partial(y \rho C_p v T)}{\partial y} = & \frac{\partial}{\partial x} \left(\lambda \frac{\partial T}{\partial x} \right) + \frac{1}{y} \frac{\partial}{\partial y} \left(y \lambda \frac{\partial T}{\partial y} \right) \\ & + q \end{aligned} \quad (9)$$

Species transport equation [18]:

$$\begin{aligned} \frac{\partial(\rho u Y_I)}{\partial x} + \frac{1}{y} \frac{\partial(y \rho v Y_I)}{\partial y} = & \frac{\partial}{\partial x} \left(\rho D_{I,m} \frac{\partial Y_I}{\partial x} \right) \\ & + \frac{1}{y} \frac{\partial}{\partial y} \left(y \rho D_{I,m} \frac{\partial Y_I}{\partial y} \right) + S_m \end{aligned} \quad (10)$$

The energy equation applied to the solid components of the computational domain reduces to a heat conduction equation since zero velocity is assigned there. Heat generation rates are introduced into the source terms of Eq. (9).

3.2 Boundary conditions

Simulations are performed using the FLUENT [19] code. The boundary conditions for the momentum, heat and mass conservation equations are as follows. On the surface of wall, uniform heat flux is utilized to indicate heat source. The model is set to “symmetric” to account for the symmetrical computational domain. The wall is specified as “heat flux” with 500 W/cm². Interior side of the wall is set as “coupled wall”. The velocity of inlet-1 and inlet-2 is set to 2 and 10 m/s,

Fig. 2 The symmetrical geometric plane structure of the reactor

respectively. A “pressure outlet” boundary condition is set at the outlet. The reactants adopted are activated carbon particles and mixture of $\text{CO}_2\text{--H}_2\text{O}$ (g), respectively. In the mixture, water vapor is used as inert gas, and mass fraction of carbon dioxide is 0.4. Activated carbon particles enter the reactor from inlet-1, while the mixture enters from inlet-2. Three thin pieces of sheets are set for the reactants to mix each other better and at the same time, the model is divided into four parts. And then pressure-based solver is selected. Through calculation, flows of activated carbon particles and CO_2 gas belong to turbulent flow. Therefore, k-epsilon (2 eqn) standard turbulent model is adopted and then standard wall surface function is used to simulate gas flow. All the above governing equations are discretised by using the finite volume approach and the SIMPLE algorithm is adopted to treat the coupling of the velocity and pressure fields. The first order upwind scheme is selected for the solution to the momentum and energy equations.

3.3 Results and discussion

3.3.1 Heat and mass transfer under high heat flux

The minimum temperature is 973.9 K when C– CO_2 reaction begins [14]. Temperature in the reactor is about 1,500 K before reaction begins, which means the reaction can occur. Figure 3a refers to the temperature field after C– CO_2 endothermic reaction is completed and Fig. 3b is an average temperature distribution graph corresponding to

Fig. 3a. Figure 4a is concerned with CO mass fraction field and Fig. 4b is relevant to CO average mass fraction distribution graph corresponding to Fig. 4a. These figures and graphs are extracted under these conditions: velocity of CO_2 is 10 m/s, velocity of activated carbon particles is 2 m/s, and operating pressure is 101.325 kPa.

It can be seen from Fig. 3a, b that temperature in the reactor drops obviously after C– CO_2 endothermic reaction is completed. The maximum temperature is just about 330 K and average temperature is 222 K. Temperature is much lower, which is below average temperature at the first part of model, and this means that the purpose of lowering temperature has come true.

Heat that has been removed and CO_2 that has been used in the process can be calculated. Mixture of CO_2 and water vapor is cooled from 1,500 to 222 K, according to the endothermic equation below:

$$Q = C_p m \Delta T \quad (11)$$

Q means endothermic or exothermic. In the case, C_p is a equivalent value of $C_{p\text{CO}_2}$ and $C_{p\text{H}_2\text{O}(l)}$, 1,427.185 J/(kg K); m refers to the mass of mixture: 0.333 kg; ΔT stands for temperature change: 1,278 K. Therefore, heat that has been removed is 607.37 kJ; molar number of CO_2 is 3.52. If the same amount of water is used to remove the heat by water phase change, only 235.9 kJ heat will be removed, which means mixture of CO_2 and water vapor will drop to 1,003.6 K.

It can be found from Figs. 3 and 4 that temperature field and CO mass fraction field have the same distribution trend:

Fig. 3 **a** Contours of temperature and **b** the distribution graph of average temperature with position on the symmetry plane of the reactor

Fig. 4 **a** Contours of CO mass fraction and **b** the distribution graph of average CO mass fraction with position on the symmetry plane of the reactor

In the first part of model, CO mass fraction is highest and temperature is lowest; in other parts, CO mass fraction is lower and temperature is higher than that of the first part. Because the inlets of reactants are in the first part, then the amount of CO_2 gas and activated carbon particles is larger, which makes them mix more fully and react more completely. CO_2 gas and activated carbon particles need to be filled into the area constantly to keep the reaction moving, so that the amount of CO_2 gas and activated carbon particles flowing into the rest parts is less and less. Later, the less the amount is, the more slightly the reaction is, so the temperature does not drop so low as that of the first part does. It can be seen from Fig. 4 that CO mass fraction is almost 0 in the fourth part of model, corresponding to Fig. 3, there is a small high temperature area in the fourth part. In spite of the fact that except the first part of the model, the reaction proceeds more and more slowly and slightly in other parts. Because of the drastic endothermic reaction in the first part, heat from the rest parts transfers to this part, which results in the drop of temperature in other parts.

3.3.2 Instantaneous heat removal

Through the calculation above, 607.37 kJ heat need to be removed for cooling mixture of CO_2 and water vapor from 1,500 to 222 K by employing C- CO_2 endothermic reaction, 3.52 mol CO_2 will be consumed in the process. The results of numerical simulation show that average mass fraction of CO_2 reaches 8.1×10^{-5} at the end of reaction. In comparison with the mass fraction 0.4 prior to reaction, it is insignificant. So it can be considered that CO_2 reacts

completely. From Figs. 3 and 4, it is known that after CO_2 gas and activated carbon particles enter the reactor, they react at a high temperature condition. According to the data of numerical simulation, time for removing 500 W/cm^2 heat at temperature of 1,500 K needs 0.142 s. The result proves that the destination of instantaneous heat removal can be realised if C- CO_2 endothermic reaction is used, because the temperature in the reactor drops to such a low level during such a short time.

3.3.3 Validation of numerical study

In the part of theoretical calculation, heat to be removed was calculated when temperature dropped from 1,500 to 300 K by cooling by means of C- CO_2 endothermic reaction, which was 470.35 kJ. And in the numerical simulation, temperature was cooled from 1,500 to 222 K, heat to be removed was 607.37 kJ. Compared with the theoretical results, it is reasonable and receivable. Table 2 shows time for removing 500 W/cm^2 heat at different temperatures. When temperatures are 1,473 and 1,573 K, it takes 0.17 and 0.09 s, respectively. In the numerical simulation, the highest temperature in the model was 1,500 K, and the reaction time could be calculated, 0.142 s, which agreed with the law of temperature and time.

4 Conclusion

In this study, C- CO_2 chemical endothermic reaction process is studied by means of theoretical calculation and

numerical simulation. Theoretical calculation results show that C–CO₂ chemical endothermic reaction can possibly remove heat enormously and instantaneously under a high heat flux density situation. The results of numerical simulation agree with theoretical calculation. Both of the results show that C–CO₂ chemical endothermic reaction has stronger heat absorption capacity than water phase change (about 2.57 times) does, which is a typical physical heat sink. It takes less time to remove the same heat. In other words, using a small amount of C and CO₂ can have the effect of instantaneous and large heat removal, instead of using a large amount of water. At the same time, the product CO can be used as supplementary medium of power source, providing energy by combustion exothermic reaction. CO₂, caused by the exothermic reaction, can also be used as a reactant of endothermic reaction. Thus the above process can form a material cycle. In this way, instantaneous heat removal and a material cycle are realized simultaneously.

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References

- Chen X (1998) Erosion on solid electrode surface caused by high density electric arc effect. *High Volt Apparatus* 4:48–50
- Luo M, Jiang J, Chang A, Zhao D (2004) Study on mechanism of electrode erosion of high-power gas spark gap switch. *High Power Laser Part Beams* 16(6):781–786
- Han Y, Wang X, Qiu L, Yu M, Zhao S, Jia W, Ai S (2010) Numerical study on heat transfer characteristics of jet array impingement. *Turbine Technol* 52(3):161–164
- Tian C, Xu H, Cao H, Si C (2009) Cooling technology for high-power solid-state laser. *Chin J Lasers* 36(7):1686–1690
- He F, Mi Z, Sun H (2006) Improvement of heat sink of endothermic hydrocarbon fuels. *Prog Chem* 18(7/8):1041–1047
- Huang M (2006) Thermal degine of the forced air-cooling radar cabinet. *Electro-Mech Eng* 22(2):12–13
- Xiao B-Q et al (2009) Mathematical analysis of pool boiling heat transfer. *Acta Phys Sin* 58(4):2523–2527
- Yang J, Chou LC, Pais MR (1996) Nucleate boiling heat transfer in spray cooling. *J Heat Transf* 118:668–671
- Bland RB et al. (1962) US3067594, Cooling with endothermic chemical reactions
- Wessberg S, Rasmussen JN (2007) (Carlsberg Brewery Co., Ltd.). CN200780024566.6, Chemical cooling
- Guo H-b (J&R Fire Fighting Group). (2010) CN201010013619.8, Chemical coolant for hot aerosol catalyst and preparation method
- Liu J, Shao J, Liu C, Rao H, Li Z, Li X (2010) Pyrolysis mechanism of hydrocarbon fuels and kinetic modeling. *Acta Chim Sin* 68(3):239–245
- Karaca F et al (2002) Economic analysis and comparison of chemical heat pump systems. *Appl Therm Eng* 22:1789–1799
- Li J, Lu K, Wang J, Wang P (2008) Influence of H₂O–CO₂ gas mixture on coke degradation. *J Anhui Univ Technol* 25(3):233–236
- Chen Z (1984) Study on kinetic and mechanism of CO₂–C reaction. *Tech Equip Environ Pollut Control* 5(1):45–49
- Cen K, Yao Q, Cao X et al (1997) Theory and application of combustion, flow, heat transfer, gasification of coal slurry. Zhe Jiang University Press, Hangzhou
- Patankar SV (1980) Numerical heat transfer and fluid flow. McGraw-Hill, New York
- Li P-W, Chyu MK (2003) Simulation of the chemical/electrochemical reactions and heat/mass transfer for a tubular SOFC in a stack. *J Power Sources* 124:487–498
- FLUENT 6 User's Guide, Fluent Inc, Lebanon, NH, 2000