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Finite element simulation of the temperature field in the large volume cubic high pressure apparatus cavity

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ABSTRACT

The finite element method is used to simulate the steady-state temperature field in diamond synthesis cell, which has a indirectly heated assembly. A series of analysis about the influence of the heating tube and pressure medium parameters on the temperature field are examined through adjusting the model parameters. The results show that temperature distribution is uneven in the sample and the highest temperature peak lies in the center position of the graphite heating tube; variation in resistivity and thermal conductivity values of graphite-pipe has little influence on the temperature distribution in the sample, thereby reducing the input power of diamond synthesis by means of improving graphite-pipe resistivity value; employing magnesium oxide as pressure medium lead to a large temperature gradient, up to 3562 K/m, and a large temperature drop of 114 K in the sample. It will lead to lower diamond quality and an energy consumption rise of diamond production. Our research would contribute to further understanding of temperature distribution mechanism in diamond synthesis cavity and bring a certain reference value on the diamond production process optimization.

1. Introduction

Since the first diamond was synthesized successfully in 1963, China has achieved great success in the abrasive grade diamond synthesis after 50 years of development. By 2012, China's annual diamond production reached to over 12 billion-carats, accounting for 90% of the world's total diamond production [1]. These achievements benefit from the two key technologies: development of large volume cubic high pressure apparatus (LV-CHPA) and indirectly heated assembly. In practice, however, these two technologies still cannot eliminate some temperature distribution issues such as overlarge temperature gradient and temperature instability existing in the cubic high press synthesis cavity during diamond synthesis process. The problem became one of the obstacles on our way to produce high-quality diamond in China. In the process of diamond synthesis by metal catalytic, the temperature is an important factor in determining the quality of diamond. Therefore, the research of temperature distribution in diamond synthesis cell is benefit to the improvement of LV-CHPA and indirectly heated assembly technology.

Diamond can be synthesized under high pressure and high temperature (HPHT) conditions. For example, Gem grade diamond can be grown in the environment of 1350 °C, 5.5 GPa [2]. This HPHT process is invisible, which bring about great difficulties for the study of the temperature distribution. Considerable efforts have been done in measuring the temperature distribution and gradient in experiments [3–6]. However, simultaneous measurement of the temperature at various points in the cell is still only theoretically possible. This restricted the improvement of diamond synthesis technology.

Numerical simulation provides a good way for us to deeply understand the physical field in the diamond cavity. Previous results [7–9] showed that the results obtained from finite element simulation of the temperature field in diamond synthesis cell have important reference value. In order to research the temperature distribution mechanism in diamond synthesis cell in the industry, we mainly did the following work: establishment of theoretical model about the diamond synthesis cavity temperature distribution by the finite element method; discussed the impact of the heating tube parameters variation on the temperature field by adjusting the model parameters. In addition, we also established a chamber temperature field model using MgO as the pressure medium, and qualitatively discussed the problems existing in using MgO as the pressure medium.

2. Computational details

2.1. Model and method

On the basis of China-type large volume cubic high-pressure apparatus (LV-CHPA), a simplified three-dimensional model was

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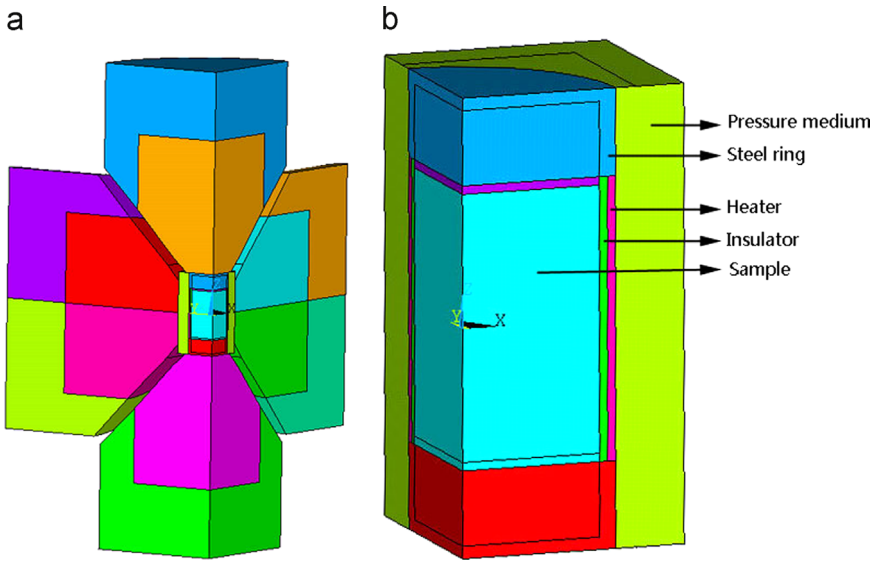

Fig. 1. Geometric model of simplified 1/4 pressure apparatus: (a) overall assembly and (b) diamond synthesis cell.

Table 1
Model dimensions and input parameters.

Components	Dimensions (mm)	Physical properties			
		κ(W/(m K))	c (J/(kg K))	ρ (kg/mm^3)	σ (Ω m)
Anvil	–	160	200	14,700	5.83×10^{-7}
Supporting ring	–	59	450	7900	9.71×10^{-8}
Steel ring	10 × (Φ34–Φ38)	59	450	7900	9.71×10^{-8}
Heater	34 × (Φ36–Φ38)	200	710	2200	8×10^{-6}
Insulator	2 × Φ34	4.19	900	2950	1×10^{12}
		33	1000	3580	1.01×10^{6}
Sample	32 × Φ38	110	585	4850	0.5×10^{-5}

established as shown in Fig. 1. The parameters of the components are listed in Table 1. Owing to the symmetry of the chamber, the simulation was carried out on the 1/4 model. Electrothermal coupling unit SOLID69 was used in the model for mapping mesh. We investigated the dependence of simulation results on element size. When the mesh size is less than 2 mm, changes in the calculation results come to be less than 5 K. So we choose the grid size of 2 mm for cavity, sample and anvils. The simulation is a steady-state thermal analysis, ignoring the process and time of thermal equilibrium. We also ignore the influence of sample deformation. Simplified boundary conditions: the boundary temperature of 308 K; the circulating water temperature of 323 K. Heating power is supplied using direct current.

2.2. Parameters

Physical properties of material under ambient conditions are shown in Table 1. The typical electrical resistivity of graphite is 8×10^{-6} Ω m at ambient conditions. Under the experimental conditions (5.5 GPa, 1573 K), the resistivity of graphite will not change greatly [10,11], so the impact of temperature and pressure on the resistivity of graphite was ignored. Graphite has a high thermal conductivity of 200 W/(m K) at ambient condition, which is matched with its low resistivity. Impact of graphite resistivity and thermal conductivity values change on temperature field will be discussed later.

Our experimental results have demonstrated that the thermal conductivity of pressure medium has an important influence on temperature field in diamond synthesis cavity. Here we summarize the thermal conductivity of the pressure transmission medium (pyrophyllite and magnesium oxide) under high pressure and high temperature (HPHT) conditions. At room temperature and atmospheric pressure, thermal conductivity of pyrophyllite is 4.19 W/(m K) [12] to 5.44 W/(m K) [13] and it will reduce with the rise of temperature. Moreover, high temperature will destroy the anisotropy of the crystal. When heated to 1300 °C, pyrophyllite thermal conductivity was approximately 1.26 W/(m K) [12]. Pressure would lead to higher density of pyrophyllite and thereby increasing its thermal conductivity. According to Ref. [13], pyrophyllite thermal conductivity has an increase of 0.12 W/(m K) per GPa in the range of 0–5.5 GPa. It is concluded that, we have a thermal conductivity value of 1.9 W/(m K) under the experimental condition. This value is close to the estimated value by Sun et al. [14].

Magnesium oxide has a high thermal conductivity of 33 W/(m K) [15] under ambient conditions. The thermal conductivity was extrapolated to 5.5 GPa, 1350 K using the following equation:

$$\kappa(P,T)=\kappa(298)(298/T)^{a}(1+K_0'P/K_0) \quad (1)$$

where $\kappa(P,T)$ is the thermal conductivity at pressure P and temperature T;$\kappa(298)$ is thermal conductivity at ambient conditions; α is the fitting parameter and 0.9 for MgO; K_0' is the derivative of bulk modulus to pressure and 4–5 for MgO, we take 5; K_0 is zero pressure bulk modulus and 131.6 [11]. Calculated by this state formula, the thermal conductivity under the experimental conditions (5.5 GPa, 1573 K) is 8.77, which is still much larger than pyrophyllite of 1.9 W/(m K).