Postprint: Combustion Characteristics in the Growth Chamber for Single Crystal Preparation by Flame Fusion Method

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Abstract

Based on the combustion of hydrogen and oxygen, the combustion characteristics within the growth chamber during the flame fusion growth of single crystals were investigated. The results indicate that: under conditions of O2 and H2 flow rates of 6 L/min and 20 L/min, respectively, the maximum temperature at the furnace center reaches 3504.3 K; based on this result, either the crystal growth position can be determined or the optimal flow rate required for crystal growth at a known position can be established. With increasing H2 flow rate, both the central and radial temperatures in the growth chamber gradually increase, while the position of the maximum temperature gradually shifts downward as the O2 flow rate increases; specifically, for each 1 L/min increase in the central O2 flow rate, the position of the maximum temperature moves downward by 5 mm, and the temperature at a location 110 mm from the nozzle rises by an average of approximately 230°C. The diameter of the H2 distribution circle has minimal influence on the central and radial temperature distributions within the growth chamber, the O2 jet penetration depth, and the ignition position.

Full Text

Combustion Characteristics in the Growth Chamber for Single Crystal Preparation by the Flame Fusion Method

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ABSTRACT

Combustion characteristics in the growth chamber for preparation of single crystal with hydro-oxygen flame fusion method were investigated. The results show that along the centerline of growth chamber a peak temperature of 3504.3 K could be reached when the flow rates of oxygen and hydrogen were 6 L/min and 20 L/min, respectively. With increasing hydrogen flow rate, the temperature at the center and along the radial direction increased gradually in the chamber. With increasing oxygen flow rate, the position of the peak temperature gradually moved downward by 5 mm when the central oxygen flow rate increased by 1 L/min, while the average temperature rose approximately 230℃ at a distance of 110 mm from the nozzle. The diameter of the hydrogen distribution circle had little effect on the center and radial temperature distributions, the central oxygen impact depth, and the ignition position of the flame. As a result, the optimal position for single crystal growth could be easily determined for preset flow rates of the two gases, and vice versa.

KEY WORDS: synthesizing and processing technic, flame fusion method, single crystal, growth chamber, combustion characteristic

Introduction

The flame fusion method is an important technique for growing high-melting-point oxide crystals. It can be used not only to prepare gemstone crystals such as ruby, sapphire, spinel, and strontium titanate, but also to prepare laser crystals like magnetoplumbite-type aluminate and optical crystals such as rutile [1-5]. During crystal growth, parameters including temperature distribution, growth rate, and especially gas composition in the growth chamber determine crystal formation, quality, and final dimensions. These parameters are related to furnace structure and burner configuration, and they continuously change and interact during the growth process [6-9]. Measurements show that the burner creates large longitudinal and transverse temperature gradients in the growth chamber. Large longitudinal temperature gradients result in high crystal cooling rates, poor crystal integrity, and high brittleness, while large transverse temperature gradients limit crystal diameter. Therefore, controlling the combustion process and temperature distribution, particularly the longitudinal and transverse atmosphere and temperature fields at the growth interface, is crucial for single crystal growth in the flame fusion method, yet experimental analysis of the combustion process and temperature distribution is difficult [10, 11]. Thus, numerical analysis is essential and necessary to theoretically investigate the combustion process and temperature distribution in the growth chamber.

This study examines the effects of gas flow rates, furnace structure, and nozzle dimensions on gas composition and flow field, as well as temperature distribution in the growth chamber, based on hydrogen-oxygen combustion.

1. Numerical Simulation Method

1.1 Furnace and Nozzle Structure Models The furnace and nozzle structures are shown in [Figure 1: see original paper]. Figure 1a presents a sectional view of the furnace structure, with a height of 250 mm and an outer diameter of 180 mm. The furnace chamber has a conical shape, with an upper diameter of 40 mm and a lower diameter of 60 mm. The refractory layer inside the furnace is made of a mixture of corundum powder and clay, the insulation layer is filled with asbestos, and the outer wall is a cylinder made of 1 mm thick steel plate. Figure 1b shows a top view of the nozzle structure, with a central hole diameter of 4 mm for oxygen and oxide powder injection, and 12 hydrogen holes of 2 mm diameter uniformly distributed on a circle with a diameter of 18 mm. Based on these structural features and considering model symmetry, the calculation model shown in [Figure 2: see original paper] was selected.

1.2 Governing Equations and Boundary Conditions The FLUENT software, based on the finite volume method with fully unstructured grids and featuring gradient algorithms based on grid nodes and grid cells, is commonly used for CFD calculations of convective heat transfer and combustion problems. According to the calculation region shown in [Figure 2: see original paper], the combustion characteristics of hydrogen and oxygen in the growth chamber mainly result from the coupling of chemical combustion reactions, turbulent flow, and convective and radiative heat transfer phenomena. The equations describing fluid flow mainly include:

Mass Conservation Equation:

$$\frac{\partial \rho}{\partial t} + \frac{\partial (\rho u_i)}{\partial x_i} = 0$$

where is density and u is the velocity vector.

Momentum Conservation Equation:

$$\frac{\partial (\rho u_i u_j)}{\partial x_j} = -\frac{\partial p}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j}$$

where τ is the stress tensor and p is pressure, defined as:

$$\tau_{ij} = \mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \mu \frac{\partial u_k}{\partial x_k} \delta_{ij}$$

where is dynamic viscosity.

Energy Conservation Equation:

$$\frac{\partial(\rho E)}{\partial t}+\frac{\partial(\rho u_i E)}{\partial x_i}=\frac{\partial}{\partial x_i}\left(k_{\text{eff}}\frac{\partial T}{\partial x_i}\right)-\sum_{j'}\frac{\partial(h_{j'}J_{j'})}{\partial x_i}+\tau_{ij}\frac{\partial u_i}{\partial x_j}+S_h$$

where E is total energy, k ff is effective thermal conductivity, J is the diffusion flux of component j , and S is the source term for chemical reaction heat.

The standard k- model was selected as the turbulence model for fluid flow, with turbulent kinetic energy k and dissipation rate equations as follows [12]:

$$\frac{\partial(\rho k)}{\partial t}+\frac{\partial(\rho k u_i)}{\partial x_i}=\frac{\partial}{\partial x_i}\left(\mu_t\frac{\partial k}{\partial x_i}\right)+G_k+G_b-\rho\varepsilon$$

$$\frac{\partial(\rho\varepsilon)}{\partial t}+\frac{\partial(\rho\varepsilon u_i)}{\partial x_i}=\frac{\partial}{\partial x_i}\left(\mu_t\frac{\partial\varepsilon}{\partial x_i}\right)+C_{1\varepsilon}\frac{\varepsilon}{k}(G_k+C_{3\varepsilon}G_b)-C_{2\varepsilon}\rho\frac{\varepsilon^2}{k}$$

where is turbulent viscosity, G is turbulent kinetic energy generated by mean velocity gradient, and G is turbulent kinetic energy generated by buoyancy.

For the H_2 and O_2 combustion reaction, since the combustion reaction rate is very high, the convective mixing of H_2 and O_2 formed by turbulence in the combustion zone is much slower than the combustion process itself. Therefore, H_2 and O_2 are quickly consumed in this zone, and the overall chemical reaction rate is controlled by turbulent mixing. In FLUENT, the eddy dissipation model of Magnussen and Hjertager is used, and the chemical reaction rate can be expressed as:

$$R_{i,\text{eddy}}=A\cdot B\cdot\rho\cdot\frac{\varepsilon}{k}\cdot\min\left(Y_{\text{fuel}},\frac{Y_{\text{ox}}}{s},B\cdot\frac{Y_{\text{pr}}}{1+s}\right)$$

where Y is the product mass percentage, and A and B are empirical constants, generally 4.0 and 0.5, respectively.

Boundary Conditions: 1. The nozzle inlets for central O_2 and H_2 were set as volume flow rates of q_1 = 6 L/min and q_2 = 20 L/min, respectively. 2. The heat flux at the upper part of the growth chamber is very small and can be neglected, thus treated as adiabatic boundary (q_1 = 0). 3. Since heat transfer in the axial direction of the furnace is much smaller than in the radial direction, axial heat transfer can be neglected, meaning the heat flux from the inner wall of the growth chamber to the outside is q. 4. The outlet pressure of the growth chamber is 0.

2. Results and Discussion

2.1 Temperature and Atmosphere Distribution Characteristics in the Growth Chamber

Based on experimental conditions, the central O_2 and H_2

flow rates were set at 6 L/min and 20 L/min, respectively. Using the above equations and boundary conditions, the FLUENT software was used to calculate the temperature and gas composition distributions in the growth chamber. [Figure 3: see original paper] shows the temperature distribution on the longitudinal section of the growth chamber. The flame exhibits central symmetric distribution in the growth chamber. Because the central O_2 flow rate is relatively large and the central hole cross-sectional area is small, a strong O_2 stream forms along the central axis and diffuses outward. According to the eddy dissipation model, O_2 mixes and combusts with surrounding H_2. Since the growth chamber is in a hydrogen-rich state, the O_2 stream is gradually consumed, forming a high-temperature flame region centered on the axis with a maximum temperature of 3504.3 K. Combined with [Figure 4: see original paper], analysis shows that the highest temperature occurs at 85.8 mm from the nozzle. By analyzing the temperature distribution characteristics along the centerline, the crystal growth position can be accurately adjusted according to the melting point temperature of the prepared crystal. For example, strontium titanate single crystal with a melting point of 2323 K should be grown at a position 125 mm from the nozzle. Meanwhile, based on the existing observation hole position, the optimal temperature required for crystal growth can be obtained by adjusting the H_2 and O_2 flow rates.

[Figure 5: see original paper] presents the temperature distribution along the inner wall of the growth chamber in the height direction. The high-temperature zone on the inner wall is mainly distributed between 60-100 mm from the nozzle. This is because the flame’ s highest temperature is at 85.8 mm from the nozzle, and the high-temperature flame radiates to the growth chamber wall, causing a high wall temperature in the symmetrical region above and below this position, with a width of approximately 40 mm. Analysis of the inner wall temperature distribution is important because while the furnace wall receives thermal radiation from the high-temperature flame, it also radiates heat to the crystal inside the growth chamber, affecting the crystal growth process. [Figure 5: see original paper] also shows that the temperature of the furnace inner wall is very high from the furnace inlet to a height of 130 mm below it. The refractory layer in this region requires high refractoriness, so the mass fraction of corundum powder in this area should be increased to enhance furnace service life.

The selection of observation hole position is critical in the flame fusion crystal growth process, as different positions have different temperatures that lead to different crystal growth states. To analyze the influence of radial temperature gradients at different heights in the growth chamber on crystal growth, [Figure 6: see original paper] shows the radial temperature distribution at different positions from the nozzle near the observation hole. At different heights, the temperature gradually decreases from the center toward the furnace wall, and the radial temperature gradient is larger at heights closer to the nozzle. Only within 10 mm from the centerline does the temperature vary significantly at different heights, while within 10-20 mm from the centerline, temperature variation is relatively small at different heights. This indicates that when the crystal diameter is less than 20 mm, the position of the crystal growth interface in the observation hole is critical, whereas when the crystal diameter exceeds 30 mm, the interface position has less influence on the crystal state. Strontium titanate crystal with a melting point of 2323 K is suitable for growth at a position 120 mm from the nozzle, which is conducive to obtaining a stable crystal growth state. Under these H_2 and O_2 flow conditions, the crystal diameter can reach approximately 10 mm.

The atmosphere field in the growth chamber is a key condition for stable single crystal growth. For example, rutile single crystals generally grow in an oxidizing atmosphere, while strontium titanate single crystals require a reducing atmosphere for stable growth. Therefore, numerical analysis is needed to obtain the atmosphere field near the crystal growth interface. [Figure 7: see original paper] shows the gas composition distribution characteristics in the growth chamber. Under the calculated hydrogen-rich condition, oxygen entering from the inlet is completely consumed by combustion with hydrogen, while hydrogen at the outlet reacts with oxygen from the air and burns out. Therefore, hydrogen and oxygen distributions in the growth chamber are mainly concentrated near the nozzle, while the atmosphere near the crystal growth position is mainly composed of water vapor and excess hydrogen. The produced water vapor is mainly distributed below the combustion zone.

2.2 Effect of H_2 Flow Rate In the flame fusion crystal growth process, the relative flow rates of H_2 and O_2 not only determine the oxidizing or reducing atmosphere in the growth chamber but, more importantly, affect the temperature distribution. Moreover, as the crystal diameter increases, the required amounts of H_2 and O_2 gradually increase. [Figure 8: see original paper] and [Figure 9: see original paper] show the effects of different H_2 flow rates on the center temperature and radial temperature distribution in the growth chamber at a constant O_2 flow rate of 9 L/min. As shown in [Figure 9: see original paper], when the O_2 flow rate remains constant, both the center axis temperature and radial temperature in the growth chamber gradually increase with increasing H_2 flow rate. Analysis of [Figure 8: see original paper] reveals that within 85 mm from the nozzle, the center temperature remains basically unchanged with increasing H_2 flow rate, indicating that this range represents the O_2 penetration depth, where temperature distribution mainly depends on the O_2 flow rate. Additionally, when the H_2 flow rate increases from 16 L/min to 17 L/min and 18 L/min, the center average temperature increases significantly. However, when the H_2 flow rate is 19 L/min and 20 L/min, the center temperature only increases at the highest position, remaining basically unchanged at other positions, indicating that excess H_2 flow at the flame tail is used to maintain a reducing atmosphere. These results show that not all O_2 at the center and its surrounding H_2 in the growth chamber actually participate in the combustion reaction. At H_2 flow rates of 16 L/min and 17 L/min, since central O_2 is in excess, the heat released by the combustion reaction mainly depends on the H_2 flow rate. The excess O_2 blows the high-temperature flue gas to deeper positions and carries away some heat, causing the temperature to decrease more rapidly at positions 90-140 mm along the centerline. At H_2 flow rates of 19 L/min and 20 L/min, theoretically, when H_2 is in excess, the heat released by the combustion reaction should depend on the O_2 flow rate, and the temperature should remain basically unchanged. However, in reality, not all central O_2 participates in the reaction. Only when the H_2 flow rate is 20 L/min does more O_2 participate in the combustion reaction, releasing more heat, increasing the temperature at the high-temperature position, while the increase in O_2 penetration depth is slightly reduced.

[Figure 9: see original paper] shows the radial temperature distribution at the observation hole position (110 mm from the nozzle) under different H_2 flow rate conditions. When the H_2 flow rate is 17 L/min and 18 L/min, the radial temperature gradually decreases from the center to the wall, but the average radial temperature increases. Only when the H_2 flow rate increases to 19 L/min and 20 L/min does the temperature increase within 8 mm from the centerline, while the temperature beyond 10 mm from the centerline remains basically unchanged or even decreases with increasing H_2 flow rate. This result indicates that larger H_2 flow rates produce more excess H_2 around the flame, and the low-temperature H_2 carries away more heat, reducing the temperature in the flame periphery. Therefore, to grow larger single crystals, the H_2 flow rate should not be excessively high.

2.3 Effect of O_2 Flow Rate [Figure 10: see original paper] and [Figure 11: see original paper] show the effects of O_2 flow rate on the center temperature and radial temperature distribution in the growth chamber at a constant H_2 flow rate of 20 L/min. As shown in [Figure 10: see original paper], when the H_2 flow rate remains constant, the trend of center temperature change in the growth chamber is basically the same with increasing O_2 flow rate. Within the 30 mm O_2 impact depth, the center temperature is basically the inlet temperature of O_2, after which O_2 combusts with surrounding H_2 to reach the maximum temperature. Under hydrogen-rich conditions in the growth chamber, the maximum temperature mainly depends on the O_2 flow rate, so the center maximum temperature gradually increases with increasing O_2 flow rate. However, not all H_2 around the O_2 participates in the reaction. When the O_2 flow rate exceeds 7 L/min, the H_2 diffusing to the O_2 surroundings is insufficient to consume all O_2, and the flame is cooled by excess O_2, causing the maximum temperature to decrease slightly. The position of the center maximum temperature gradually moves downward with increasing O_2 flow rate, moving down by 5 mm for each 1 L/min increase in central O_2 flow rate.

[Figure 11: see original paper] shows that at a constant H_2 flow rate, the radial temperature distribution trends at the observation hole position are basically the same under different O_2 flow rate conditions, with radial temperature gradually decreasing as radius increases, and larger central O_2 flow rates producing larger radial temperature gradients. Under hydrogen-rich conditions in the growth chamber, each 1 L/min increase in central O_2 flow rate raises the temperature at the observation hole position (110 mm from the nozzle) by an average of 230℃. Therefore, to grow larger single crystals, there must be a reasonable H_2 and O_2 flow rate, and as the crystal continues to grow and its diameter increases, the required O_2 flow rate also increases continuously.

2.4 Effect of H_2 Distribution Circle Diameter [Figure 12: see original paper] and [Figure 13: see original paper] show the effects of different H_2 hole distribution circle diameters on the center temperature and radial temperature distribution in the growth chamber at constant H_2 and O_2 flow rates of 20 L/min and 9 L/min, respectively. As shown in [Figure 12: see original paper] and [Figure 13: see original paper], when the H_2 and O_2 flow rates remain constant, the H_2 distribution circle diameter has little effect on the center temperature distribution and radial temperature distribution in the growth chamber. The calculation results indicate that the H_2 distribution circle diameter has minimal influence on O_2 impact depth and ignition position. This is because the combustion reaction rate of H_2 and O_2 is very high, and H_2 and O_2 diffuse to the combustion reaction interface very quickly. The temperature distribution in the growth chamber mainly depends on the O_2 flow rate and H_2 flow rate.

Conclusions

1. Under conditions of O_2 and H_2 flow rates of 6 L/min and 20 L/min, respectively, the maximum temperature at the furnace center is 3504.3 K. The center temperature at 125 mm from the nozzle can reach the melting point temperature of strontium titanate crystal (2323 K), allowing determination of the crystal growth position under these conditions. Alternatively, based on existing observation hole positions, the optimal temperature required for crystal growth can be obtained by adjusting the H_2 and O_2 flow rates.
2. When the O_2 flow rate remains constant, both the center and radial temperatures in the growth chamber gradually increase with increasing H_2 flow rate, while the increase in O_2 penetration depth is slightly reduced.
3. When the H_2 flow rate remains constant, the position of the center maximum temperature gradually moves downward with increasing O_2 flow rate, moving down by 5 mm for each 1 L/min increase in central O_2 flow rate, raising the temperature at the observation hole position (110 mm from the nozzle) by an average of approximately 230℃.
4. When the H_2 and O_2 flow rates remain constant, the H_2 distribution circle diameter has minimal effect on the center and radial temperature distributions, O_2 impact depth, and ignition position.

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