Effect of Al(OH)₃ addition on densification mechanism and properties of reaction-sintered mullite-cordierite composite ceramics

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ABSTRACT
Mullite composite ceramics were fabricated by using mullite powder from waste coal gangue and Al(OH)₃ as starting materials. The effects of sintering temperature and Al(OH)₃ content on phase composition, microstructure, and mechanical properties of the ceramics were systematically investigated. Results show that the bulk density and flexural strength of composite ceramics increase as the temperature increases from 1480°C to 1560°C. The composite ceramics exhibit optimal performances with addition of 10 wt.% Al(OH)₃ at 1560°C, a bulk density of 2.43 g/cm³ and a flexural strength of 124.28 MPa, respectively. Moreover, additional Al(OH)₃ promotes a reaction between SiO₂ and Al₂O₃ and forms more mullite phase. The increase in mullite content endows the composite ceramics with high mechanical properties. Scanning electron microscope images indicate that the mullite particles exhibit an interlocking structure, while the corundum phase is “pinned” within the mullite interlocking structure, contributing to the mechanical properties of composite ceramics.

1. Introduction
Coal gangue is a complex solid waste material generated during coal mining. Statistical analysis reveals that coal gangue production accounts for 15–20% of raw coal production [1–3]. The total amount of accumulated coal gangue is approximately 6 billion tons in China [4]. Carbon residues and sulfides in coal gangue pose a significant environmental threat [5,6]. The traditional recycling methods of coal gangue are based on landfill, power generation and mineral recovery, which also cause environmental pollution [7–9]. The utilization of value-added coal gangue benefits sustainability, energy saving, and environmental enhancement, so related studies have attracted more attention. The main components of coal gangue are Al₂O₃ and SiO₂. The design and preparation of high-performance ceramics by coal gangue is an attractive strategy. Quartz is formed during heat treatment of coal gangue, which easily transforms into a glass phase above 1500°C [10]. The presence of a glassy phase affects the mechanical properties of mullite. It is reported that the addition of alumina source promotes the reaction between alumina and excess quartz present in coal gangue [4,7,11]. It can reduce the formation of the glassy phase and significantly improve the mechanical properties. Many studies have reported that coal gangue can be used as a raw material to prepare innovative mullite [12–14].

Mullite is a nonstoichiometric compound whose composition generally lies between 3Al₂O₃ - 2SiO₂ and 2Al₂O₃·SiO₂. It possesses the advantages of high chemical stability, low thermal expansion coefficient, excellent chemical resistance, superior thermal shock resistance and good creep resistance [15–17]. Thus, preparing mullite ceramics from coal gangue is essential for developing value-added products. Some researchers have attempted to use coal gangue to replace traditionally pure materials during the fabrication of mullite-based ceramics to reduce costs. For example, Ji et al. [18] found that mullite ceramics could be fabricated from coal gangue by adding γ-Al₂O₃ and bauxite as aluminum sources, rendering excellent performance. Liu et al. [19] used coal gangue and high alumina waste to produce ceramics with excellent properties and realized the resource utilization of coal gangue. Hao et al. [20] reported that low-density ceramic proppants could be successfully prepared from coal gangue and flint clay.

However, the typical problem in these studies is the presence of impurities in coal gangue, which can form a liquid phase during the one-step synthesis (directly using coal gangue raw materials for synthesis by solid phase method) and lead to inferior mechanical properties [10]. Therefore, coal gangue is treated by the molten salt method to reduce impurities, as reported...
Moreover, Al(OH)₃ has been used as an Al source because of its high reaction activity for preparation of functional ceramics. For instance, Fu et al. [22] utilized waste coal fly ash and Al(OH)₃ as raw materials to fabricate mullite ceramic membranes. The results revealed that Al(OH)₃ is better than Al₂O₃ for preparing mullite whiskers, rendering better mechanical properties and a higher degree of mullitization than Al₂O₃. Also, Li et al. [23] prepared porous whisker-structured mullite using fly ash solid waste and different aluminum sources, such as Al₂O₃, Al(OH)₃, and AlF₃. They demonstrated that the mullite ceramics with Al(OH)₃ are better than those with Al₂O₃ and form an interlocking structure with a high flexural strength of 100 MPa.

Consequently, α-Al₂O₃ exerts a significant influence on the morphology and mechanical properties of ceramics. However, there are few reports on the synthesis of composite ceramics with coal gangue after decomposing Al(OH)₃ into α-Al₂O₃.

Therefore, the purpose of this work was to investigate the preparation of mullite composite ceramics with excellent mechanical properties by using molten salt and solid phase sintering methods. Besides in situ synthesis of mullite powders from industrial solid waste coal gangue as Al-Si sources, chemically pure Al(OH)₃ has also been used as the Al-source due to its high reaction activity in mullite formation. It was found that the different amounts of Al(OH)₃ significantly influenced the physical properties, phase composition, microstructure and mechanical strength of composite ceramics. Because in-situ decomposition of Al(OH)₃ can provide higher reaction activity of Al₂O₃ in the subsequent mullitization reaction.

### 2. Experimental section

#### 2.1. Raw materials

Mullite powder and Al(OH)₃ powder (Sinopharm Chemical Reagent Co. Ltd., China) were used as starting materials. The preparation method and processing conditions for mullite powder are described in our previous report [21]. The composition and particle size of starting materials are summarized in Table 1.

Figure 1(a) shows the morphology of coal gangue after molten salt treatment. It is composed of needle-shaped crystals. In addition, the crystalline phase of the sintered powder is composed of mullite and a small amount of quartz, as shown in Figure 1(b).

#### 2.2. Sample preparation

The weight ratio of mullite to Al(OH)₃ was calculated according to Table 2. First, the starting materials were weighed and mixed by a planetary ball milling machine at 300 rpm for 3 h, to form a homogeneous mixture. Then, 5 wt.% polyvinyl alcohol (PVA) solution was used as the binder. The uniform mixture was uniaxially pressed into cylindrical compacts (Ø 15 mm × 5 mm) and square compacts (40 mm × 40 mm × 6.5 mm) at 20 MPa by a manual tablet compaction machine. After demolding, the pressed compacts were dried at 110°C for 24 h. Finally, these obtained compacts were sintered at different sintering temperatures (1480–1580°C) in an electric furnace at a heating rate of 3°C·min⁻¹ for 3 h, and all samples were cooled down naturally. Five samples were tested for each batch of samples, and the average was taken.

#### 2.3. Characterization

The Archimedes principle was adopted to analyze the bulk density and apparent porosity of the composite ceramics. The water absorption was determined using a Ceramic Water Absorption Tester (Model TXY-250, Xiangtan, China). The shrinkage rate was calculated based on the sample diameter before and after sintering. A uniaxial compressive tester (WDW-50, Kaiqiangli, China).

### Table 1. The characteristics of starting materials.

| Chemical composition/wt.% | Al₂O₃ | SiO₂ | Fe₂O₃ | TiO₂ | K₂O | MgO | LOI | d₅₀/μm |
|---------------------------|-------|------|-------|------|-----|-----|-----|--------|
| Mullite powder            | 42.30 | 38.87| 1.85  | 0.65 | 1.62| 0.63| 2~17| 11.3   |
| Al(OH)₃                   | ~99.9 | -    | -     | -    | -   | -   | -   | 9.2    |

*LOI: loss on ignition.

Figure 1. The characterization of coal gangue after molten salt treatment: (a) SEM image and (b) XRD pattern.
China) was used to determine the flexural strength by a three-point bending method with a span length of 30 mm and a loading rate of 0.05 mm/min, respectively. The average of five independently measured values is reported as the flexural strength of a given composition. A thermogravimetric and differential scanning calorimeter (TG-DSC, STA449C, Netzsch Co. Ltd., Germany) was used for the thermal analysis of mixed powders in the temperature range of 25 to 1000°C at a heating rate of 10°C min\(^{-1}\). The crystalline phase compositions of as-sintered ceramics were determined using an X-ray diffractometer (XRD, Smartlab SE, Rigaku), equipped with Cu Ka radiation (\(\lambda = 1.5406 \text{ Å}\)). Herein, the phase content was calculated in accordance with the normalized reference intensity ratio (RIR). The microstructure of as-sintered ceramics was observed by a scanning electron microscope (SEM, Zeiss, Gemini Sigma 300/VP, Germany). The particle size of the starting powders was obtained by a laser granulometry analyzer (Malvern Mastersizer 2000).

### 3. Results and discussion

#### 3.1. Thermal analysis

Figure 2 shows the TG-DSC curves of the starting mixture (mullite and Al(OH)\(_3\)). As shown in Figure 2, the weight loss resulted in two endothermic peaks and two exothermic peaks in the DSC curve. The first weight loss (approximately 11\%) was attributed to physical water absorption and evaporation below 282.4°C, with an endothermic peak appearing in the DSC curve. The second weight loss (4.39\%) occurred in the temperature range of 550°C to 1024°C due to Al(OH)\(_3\) decomposition, corresponding to the appearance of an exothermic peak at 795°C. Moreover, an endothermic peak occurred at 985°C, which could be attributed to the crystalline transformation of \(\gamma\)-Al\(_2\)O\(_3\) to \(\alpha\)-Al\(_2\)O\(_3\). Another exothermic peak was observed at 1150°C, which corresponded to mullite formation due to the reaction between \(\alpha\)-Al\(_2\)O\(_3\) and SiO\(_2\) [24].

#### 3.2. Phase composition

Figure 3(a) shows the effect of 5 wt.% Al(OH)\(_3\) addition on the phase evolution of composite ceramics after sintering at different temperatures (1480–1580°C). The relative amounts of different phases were also calculated based on XRD results, as shown in Figure 3(b). Figure 3(a) shows that the main crystal phase in composite ceramics was the mullite (3Al\(_2\)O\(_3\) \(\cdot\) 2SiO\(_2\), orthorhombic system, PDF #74-2419). Moreover, corundum (Al\(_2\)O\(_3\), rhombohedral system, PDF #74-1081) existed as a minor phase and the intensity of the corresponding peak decreased at >1530°C. Interestingly, as shown in Figure 3(b), the mullite content accounted for 74.9\% after adding 5 wt.% Al(OH)\(_3\) at 1480°C. At 1500°C, the mullite content increased from 74.9 to 98.7 wt.% This is mainly because the synthesized mullite powders are impure and contain a small amount of quartz, while Al(OH)\(_3\) is converted to Al\(_2\)O\(_3\) at low temperatures. Subsequently, quartz can react with adjacent Al\(_2\)O\(_3\) particles to form secondary mullite with the increase in sintering temperature. Moreover, no remarkable change in the phase content was observed with increase in temperature from 1500°C to 1550°C. Therefore, the mullitization was almost completed at 1500°C. Equations (1–4) express the reactions during the sintering process [24]:

\[
2\text{Al(OH)}_3 \rightarrow \gamma\text{-Al}_2\text{O}_3 + 3\text{H}_2\text{O} \quad (1)
\]

\[
2\text{Al(OH)}_3 \rightarrow \rho\text{-Al}_2\text{O}_3 + 3\text{H}_2\text{O} \quad (2)
\]

![Figure 2. TG-DSC curve of the composite powders.](image-url)
Figure 3. The evolution of phase content in composite ceramics with 5 wt.% Al(OH)$_3$ after sintering at different temperatures: (a) XRD patterns and (b) phase content.

\[
\rho - \text{Al}_2\text{O}_3 (\text{or} - \text{Al}_2\text{O}_3) \rightarrow \alpha - \text{Al}_2\text{O}_3 \quad (3)
\]

\[
3\text{Al}_2\text{O}_3 + 2\text{SiO}_2 \rightarrow \text{Al}_6\text{Si}_2\text{O}_{13} \quad (4)
\]

Similar results have been reported by Liu et al. [19] and Bella et al. [24], and they have also indicated that the increase in phase content could be ascribed to the formation of secondary mullite.

XRD patterns of the composite ceramics with different amounts of Al(OH)$_3$, sintered at 1560°C, are presented in Figure 4(a), indicating the presence of both mullite and corundum phases. Figure 4(b) presents the relative fraction of the mullite content. For the composite ceramic with 5 wt.% Al(OH)$_3$, the mullite content increased remarkably, accounting for 97.9%, along with minor corundum, accounting for 2.1%. Also, the mullite content decreased with the increase in Al(OH)$_3$ content. The mullite content in samples A, B, C and D was found to be 97.9%, 95.8%, 83.1% and 81.5%, respectively. At the Al(OH)$_3$ content of 10 wt.%, the intensities of corundum peaks increased. Moreover, the further increase in Al(OH)$_3$ content to 20 wt.% enhanced the corundum content to 18.5%. As mentioned earlier, the mullite phase was completely formed above 1500°C. Therefore, the evolution of corundum phase content can be mainly attributed to Al(OH)$_3$ addition.

3.3. Physical properties of the composite ceramics

The shrinkage and water absorption of composite ceramics with different amounts of Al(OH)$_3$ in the sintering temperature range of 1480 to 1580°C are shown in Figure 5. Clearly, the variation trend of the shrinkage was opposite to that of the water absorption. With increase in sintering temperature from 1530 to 1560°C, the shrinkage became prominent because high temperature promoted sintering densification.

Moreover, the alkaline oxides (MgO, CaO, and Fe$_2$O$_3$) in the raw coal gangue easily increased the amount of liquid phase, filling the open pores and cracks and enhancing the densification of composite ceramics [25]. However, the shrinkage of the composite ceramics decreased significantly after sintering at 1580°C, which can be related to the expansion caused by

Figure 4. The phase evolution in composite ceramics, sintered at 1560°C, with respect to the Al(OH)$_3$ content: (a) XRD patterns and (b) phase content.
abnormal grain growth of mullite at high sintering temperatures.

Furthermore, Al(OH)₃ addition had a positive influence on the density of as-sintered composite ceramics, as shown in Figure 5(a). The shrinkage of composite ceramics with the addition of Al(OH)₃ (<15 wt.%) increased with increase in sintering temperature, followed by a decrease at all temperatures except for 1480°C. Compared with the addition of 10 wt.% Al(OH)₃, adding 15 wt.% Al(OH)₃ resulted in a more significant shrinkage at 1580°C. This phenomenon may be related to the mutual cancellation of the shrinkage caused by Al(OH)₃ decomposition and the expansion caused by secondary mullite formation [11,26].

The effect of sintering temperature on the sintering behavior of composite ceramics with different amounts of Al(OH)₃ is shown in Figure 6. The bulk density of the composite ceramics initially increased with the increase in sintering temperature, followed by a decrease (Figure 6(a)). On the other hand, the apparent porosity exhibited the opposite trend (Figure 6(b)). When the temperature increased from 1480 to 1580°C, the bulk density of the composite ceramics increased from 2.14 to 2.34 g/cm³, while the apparent porosity of the composite ceramics decreased from 26.52 to 0.12%. The bulk density of the composite ceramics exhibited a different trend with increase in Al(OH)₃ content. At 1560°C, the maximum bulk density of composite ceramic with 10 wt.% Al(OH)₃ was found to be 2.34 g/cm³. Consistent with the change in water absorption in Figure 6(b), the densification of the sample was higher than other samples. Combined with microstructural analysis, as shown in Figure 8(b), it can be concluded that the mullite grains were tightly bonded. This densification behavior can be attributed to two factors: (1) the content of the liquid phase increased at high temperature, closing some visible pores and aiding the densification process [3,4], and (2) Al(OH)₃ decomposed into highly active Al₂O₃ phase and the thermal decomposition shrunk the samples [27]. When the sintering temperature was >1560°C, the bulk density rapidly decreased and the apparent porosity increased. Meanwhile, the mullite grains exhibited abnormal growth. As reported elsewhere

Figure 6. The physical properties of the composite ceramics (A–D) after sintering at different temperatures: (a) bulk density and (b) apparent porosity.
abnormally grown mullite grains are responsible for weakening microstructural densification. Also, when the Al(OH)₃ content was >15 wt.%, the expansion originating from mullitization was more significant than the shrinkage of densification sintering, which reduced the density and increased the porosity of composite ceramics.

3.4. Influence of sintering temperature on microstructure

Figure 7 shows the SEM images of the composite ceramics with 10 wt.% Al(OH)₃ after sintering at elevated temperatures (1480°C to 1580°C). As shown in Figure 7, the composite ceramics possessed rod-like mullite crystals, covering the entire surface. At 1480°C, the composite ceramic exhibited a loose structure with many pores and the mullite with different amounts of elongated grains can be clearly observed (Figure 7(b)). At 1500°C, the mullite grains gradually became coarse (Figure 7(d)). When the sintering temperature increased to 1530°C, the columnar mullite grains grew rapidly along the diameter, forming thick and short columnar mullite grains (Figure 7(f)). This is because the liquid phase increased and filled the pores and grain boundaries with increasing temperature, and the corresponding diffusion rate and mass transfer rate also increased. The mass transfer through the liquid phase around the mullite grains caused rearrangement. At 1560°C, the columnar mullite crystals were better developed and the junction with the substrate was tightened (Figure 7(g)). As shown in Figure 8(h), the corundum phase migrated into the pores of mullite particles, and the mullite with a “pinned” structure was created, which contributed to the mechanical properties of the composite ceramics [29,30].

Moreover, the amount of liquid phase significantly increased after being sintered at 1580°C, as shown in Figure 7(i,j). The higher sintering temperature provided

Figure 7. SEM images of the composite ceramic with 10 wt.% Al(OH)₃ content after sintering at different temperatures: (a,b) 1480°C, (c,d) 1500°C, (e,f) 1530°C, (g,h) 1560°C, and (i,j) 1580°C.

Figure 8. SEM images of composite ceramics, sintered at 1560°C, with different amounts of Al(OH)₃: (a, A) 5 wt.%, (b, B) 10 wt.%, (c, C) 15 wt.%, and (d, D) 20 wt.%. 
a strong driving force for the growth of mullite grains. The dissolution and crystallization of mullite in the liquid phase caused abnormal growth of the mullite grains. The increased grain size and liquid phase are detrimental to the mechanical properties of composite ceramics, as reported in the literature \[3,9\].

### 3.5. Effect of Al(OH)\(_3\) content on microstructural evolution and mechanical properties

Figure 8 shows the SEM images of the composite ceramics with different amounts of Al(OH)\(_3\) after sintering at 1560°C, revealing that the Al(OH)\(_3\) content significantly influenced the morphological evolution of the as-synthesized composite ceramics. When the Al(OH)\(_3\) content was 5 wt.\%, the structure was loose with large pores (Figure 8(a,A)) that was mainly created by initial particles accumulation. A further increase in Al(OH)\(_3\) content to 10 wt.\% (Figure 8(b,B)) promoted an interlocking structure formed by columnar mullite and a small amount of corundum. Also, the tips of columnar mullite were rounded, which could effectively improve the mechanical properties. When the content of Al(OH)\(_3\) increased from 10 to 15 wt.\%, most of the columnar mullite in the composite ceramic gradually changed into a short columnar structure. The aspect ratio of samples C and D became significantly smaller than that of samples A and B. When Al(OH)\(_3\) content further increased to 20 wt.\%, the liquid reacted with the corundum phase through a peritectic reaction and the grain boundaries became blurred, as shown in Figure 8(d).

Figure 9 shows the flexural strength of composite ceramics with different amounts of Al(OH)\(_3\). The flexural strength gradually increased with increase in Al(OH)\(_3\) content from 5 to 20 wt.\%, followed by a decrease. Apparently, the flexural strength of the composite ceramics with Al(OH)\(_3\) was significantly greater than that of the ceramics without Al(OH)\(_3\). The composite ceramic with 10 wt.\% Al(OH)\(_3\) displayed a high flexural strength of 124.28 MPa, which can be ascribed to the presence of long columnar mullite grains with an interlocking structure, acting as a reinforcement phase to improve the flexural strength. This phenomenon can be explained according to Figure 8(b). When the Al(OH)\(_3\) content exceeded 10 wt.\%, the flexural strength of ceramics decreased. The flexural strength of sample D was 102.13 MPa, which might be related to the formation of microcracks and the liquid phase during the firing process. In Figure 8(c,d), the microcracks inside the samples can be clearly observed.

To highlight the advantages of composite ceramics in this work, the mechanical properties of similar materials reported in the open literature were compared (Table 3). In general, composite ceramics that are prepared using pure raw materials such as Al\(_2\)O\(_3\) and SiO\(_2\), require sintering temperatures of at least 1600°C. However, a lower sintering temperature of around 1480 to 1560°C was required in this work. Compared with high purity metal oxide, these alternative coal gangue raw materials used as the Si-Al source can be sintered at a lower temperature while exhibiting excellent mechanical strength.

### 3.6. Sintering mechanism of composite ceramics

To better explain the sintering mechanism of composite ceramics after Al(OH)\(_3\) addition, a model of the reaction sintering process was proposed. As shown in Figure 10, the reaction process can be divided into three steps:

1. **Solid-phase bonding**

   The γ-Al\(_2\)O\(_3\) phase can be transformed into α-Al\(_2\)O\(_3\) at <1000°C [34]. The crystals of γ-Al\(_2\)O\(_3\) are face-centered cubic stacks with vacancies, where oxygen

![Figure 9](Image) **Figure 9.** The flexural strength of composite ceramics with different amounts of Al(OH)\(_3\) after being sintered at 1560°C.
Table 3. Comparison between the properties of the as-fabricated mullite ceramics in this work with others reported in the literature.

| Samples                        | Raw materials                  | Sintering temperature (°C) | Sintering method   | Bulk density (g/cm³) | Flexural strength (MPa) | Ref.  |
|--------------------------------|--------------------------------|----------------------------|--------------------|----------------------|-------------------------|-------|
| Mullite composite ceramics     | Cristobalite and α-alumina     | 1650°C                     | Traditional sintering | 2.69                 | 128                     | [31]  |
| Mullite ceramics               | Solid wastes (high alumina)    | 1500°C                     | Traditional sintering | 2.52                 | 71.76                   | [19]  |
| Mullite ceramics               | Gangue and Al₂O₃               | 1400°C                     | Traditional sintering | /                    | 97.60                   | [32]  |
| Mullite composite ceramics     | Gangue and Al(OH)₃             | 1560°C                     | Traditional sintering | 2.34                 | 124.28                  | This work |
| Mullite composite ceramics     | CSK and α-Al₂O₃                | 1600°C                     | Pressureless sintering | /                    | 124.58                  | [33]  |

Figure 10. The model of reaction sintering process of composite ceramics.

atoms approximate a hexagonal close packing arrangement. Also, α-Al₂O₃ and SiO₂ particles are close to each other.

(2) Liquid-solid sintering

Liquid-solid sintering initiates at 1100°C due to the formation of glass phases from a few impurities, such as Fe₂O₃ and K₂O, in the starting powders. The contact area between mullite particles and Al₂O₃ increases, which further promotes the solid-phase reaction. γ-Al₂O₃ crystals possess a spinel structure with defects, which causes grain lattice distortion and activation [35]. Therefore, Al₂O₃ is prone to react quickly with SiO₂ to form mullite at this stage.

(3) Complete mullitization

The mullitization is complete when the temperature increases to 1500°C. In addition, the coal gangue raw material contains a certain amount of impurities, which can easily form a liquid phase during the sintering process. This increases the driving force for sintering and leads to sintering densification [36]. At this stage, mullite grains tend to grow anisotropically in the liquid phase and the acicular mullite grains are coarsened. At the same time, the mass transfer becomes dominant in the sintering process and mullite is formed with an interlocking structure. The stacked interlocking structure is conducive to the improved mechanical properties of mullite ceramics [37,38].

4. Conclusions

In this work, high-performance composite ceramics with columnar mullite crystals were successfully fabricated by solid-phase reaction at 1480–1580°C using mullite powder and Al(OH)₃ as the raw materials. The effects of sintering temperature and Al(OH)₃ content on the structural evolution and mechanical properties of the composite ceramics were studied in detail. As the sintering temperature rose, the bulk density of the composite ceramics initially increased, followed by a decrease. Most of the needlelike mullite gradually grew into short columnar mullite with interlocking structure in the temperature range from 1480 to 1560°C. Al(OH)₃, as an additive, decomposed into Al₂O₃ and reacted with quartz to form the mullite phase. The excess corundum was "pinned" at the grain boundaries, enhancing the flexural strength. The sample with 10 wt.% Al(OH)₃ addition showed excellent
properties, with the maximum bulk density of 2.34 g/cm³ and the flexural strength of 124.28 MPa, corresponding to an improvement of 36% compared with the Al(OH)₃-free composite ceramics (79.25 MPa).

Disclosure statement

No potential conflict of interest was reported by the author(s).

Funding

This work was supported by (Grant No. 202104a050200033), the Foundation of State Key Laboratory of High-efficiency Utilization of Coal and Green Chemical Engineering (Grant No. 2021-K19), the National Natural Science Foundation of China (Grant No. The National Natural Science Foundation of China 11872001), Anhui Provincial Major Science and Technology Special Program (Grant No. 18030901049), the School-enterprise Cooperation Projects (Grant No. HX2021062279), and the College Students Innovation and Entrepreneurship Training Program (Grant No. S202110361147; the Key Technologies R&D Program of Anhui Province of China

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