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### Feature article

# Preparation and properties of in-situ mullite whiskers reinforced aluminum chromium phosphate wave-transparent ceramics

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### ABSTRACT

In-situ mullite whisker reinforced aluminum chromium phosphate wave-transparent ceramics were designed and prepared. The phase transformation, microstructure, mechanical and electrical properties of the ceramics were investigated, and the mechanisms of in-situ growth and toughening were discussed. Results indicated that the in-situ growth of mullite whisker significantly improved the mechanical properties of the matrix, especially the high temperature flexural strength. The room temperature flexural strength, 1000 °C flexural strength and fracture toughness of the ceramics were 135.60 MPa, 121.71 MPa and 4.52 MPa m<sup>1/2</sup>. After sintering at 1500 °C, the optimum properties of  $\epsilon'$ ,  $\tan\delta$  and microwave transmittance at region 8–12 GHz were <3.6, <0.03 and >80%, respectively. The sinterability of ACP matrix was improved by the in-situ process of high mullitization above 1450 °C. Using ACP binder as the raw material can avoid the phase transformation from B-AlPO<sub>4</sub> to T-AlPO<sub>4</sub>. The synthesized mullite whiskers played a role in toughening by whiskers fracture, crack deflection and whisker pulling out.

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## 1. Introduction

Aluminum chromium phosphates composites have been widely used in refractory ceramics for many years because of its outstanding chemical stability, chemical compatibility and high melting point (1800 °C, higher than aluminum phosphate 1400 °C). It is also well-known that it can be used as an attractive candidate material for high temperature wave-transparent applications, due to its relatively low thermal conductivity, low dielectric constant and low density [1–5]. However, the brittleness, relatively low mechanical strength and poor sinterability [6] of it has restricted the application for high temperature structural material. Throughout the current state of phosphate-based composites researches, continuous fibers, such as silicon carbide fiber and quartz fiber were used to reinforce aluminum chromium phosphates matrix to obtain better mechanical properties [2,3,7–13]. But silicon carbide fibers must be subjected to anti-oxidation pretreatment before use, which leads to more complex process, high cost and high dielectric constant. Siliceous fiber reinforced aluminum chromium phosphates matrix are also limited in the service temperature by its low heat-resistance, low erosion resistance and low mechanical strength

after being heated. Moreover, with the escalation of flight speed and service environment, high temperature wave-transparent materials were required for more excellent high temperature mechanical properties and longer life. Therefore, in order to improve the high temperature service capacity of aluminum chromium phosphates, it is significant to explore new reinforcement and enhance process to develop this low-cost, high-performance material.

Mullite(3Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>) is the only stable phase in SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> binary system, showing a high melting point(1850 °C), high temperature creep resistance, low density(3.17 g cm<sup>-3</sup>) and low dielectric constant(6.4–7.0) [14–19]. Furthermore, the thermal expansion coefficient(CTE)(4.4–7.6 × 10<sup>-6</sup> °C<sup>-1</sup>) of it is close to aluminum phosphate (5.5–5.75 × 10<sup>-6</sup> °C<sup>-1</sup>) [20,21]. P. Robinson et al. [22] once studied the solid reactions of SiO<sub>2</sub>–Al<sub>2</sub>O<sub>3</sub>–P<sub>2</sub>O<sub>5</sub> ternary system, They concluded that this system can finally form a coexist stable structure with aluminum phosphate and mullite crystals. Thus it can be seen that these two substances not only have pleasurable thermal matching characteristics, but also have favorable chemical compatibility, which provide positive theoretical support for mullite in-situ growth.

In this paper, considering those pleasurable thermal matching, chemical compatibility, lower dielectric constant and density of mullite and aluminum phosphate, the in-situ mullite whiskers reinforced aluminum chromium phosphate matrix ceramics were firstly designed and prepared. The phase transformation,

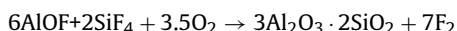
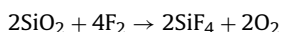
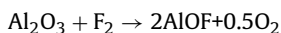
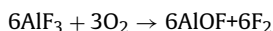
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microstructure, mechanical and electrical properties of the ceramics were investigated, and the mechanisms of in-situ growth and toughening were discussed as well.

## 2. Experimental procedure

Mullite<sub>w</sub>/aluminum chromium phosphate matrix ceramics were prepared by the raw materials consisting of aluminum chromium phosphates binder (denoted by ACP, obtained from Al(OH)<sub>3</sub>, CrO<sub>3</sub>, H<sub>3</sub>PO<sub>4</sub> and CH<sub>3</sub>OH using molar ratio at Al:Cr:P=3:1:9, as reported in Ref [23]), Al(OH)<sub>3</sub> (Kelong Chemical, AR) and fused SiO<sub>2</sub> powders (Kelong Chemical, AR). The mass ratio between aluminum chromium phosphate and mullite was kept at 3:2 through controlling the mixture ratio of raw materials. The 5 wt% AlF<sub>3</sub>·3H<sub>2</sub>O powders were added as assistant agents for in situ mullite whiskers[18,24]. Then the raw materials were mixed by wet ball milling for 24 h. After drying and sieving, the precursors were dry-pressed uniaxially into rectangular bars (6.50 mm × 6.50 mm × 40 mm and 5 mm × 5 mm × 40 mm) at 30 MPa. The green formed samples were placed in hermetic alumina crucible and sintered at various temperatures for 4 h in air.

The chemical reaction of in-situ mullite whiskers can be expressed by the following equations:



The flexural strength was evaluated by three-point bending with a span of 16 mm using a crosshead speed of 0.5 mm/min at room temperature and 1000 °C, respectively. The fracture toughness was evaluated by the single-edge pre-cracked beam (SEPB) method. The procedure of thermal shock treatment was as follows: the samples were heated to 1000 °C with 5 °C/min rate in furnace, held for 15 min and then quenched in water to obtain a matrix failure sample ( $\Delta T = 975$  °C). Vickers indentations were introduced to make the main cracks on the polished surface of the sample with an indent load of 9.81 N for 13 s.

The phase transformation of the samples sintered at various temperatures was identified by XRD analysis (X'Pert PRO, PANalytical B.V., Netherlands). The relative dielectric constants ( $\epsilon_r$ ) were measured at region 8–12 GHz by Vector Network Analyzer (E5071C, Agilent). The morphology of fractured surfaces and cracks propagation of the samples was observed by SEM (EVO-18, ZEISS, Germany and TM1000, Hitachi, Japan). The bulk densities of the samples were measured by Ceramic density tester (QL-120C, MZ, China). The mechanical properties as mentioned above were measured by Universal Testing Machine (RGM-4100, Reger, China).

## 3. Results and discussion

### 3.1. XRD analysis

The XRD patterns of precursors sintered at different temperatures from 800 °C to 1550 °C are shown in Fig. 1. At the temperature of 800 °C, no crystalline phase was observed, the ACP, Al(OH)<sub>3</sub> and fused SiO<sub>2</sub> in precursors remained amorphous. Our previous research on ACP thermal transformation[23], suggests that it doesn't crystallize until 1000 °C, which is beneficial for the ceramics to avoid phase transformation from B-AlPO<sub>4</sub> to T-AlPO<sub>4</sub> between 753 °C and 815 °C, because the transformation could induce a mass

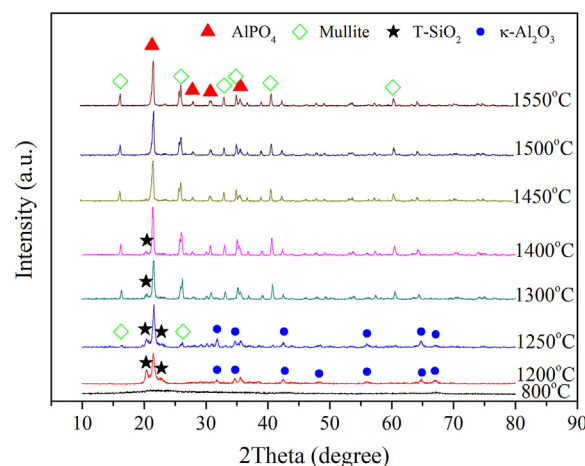


Fig. 1. XRD patterns of Mullite whisker/ACP precursors sintered at various temperatures.

of cracks in ceramic body[25]. After heating at 1200 °C, the amorphous phases began to crystallize, forming orthorhombic AlPO<sub>4</sub>, T-SiO<sub>2</sub> and  $\kappa$ -Al<sub>2</sub>O<sub>3</sub>. As temperature rose to 1250 °C, a small quantity of mullite was obtained, accompanying with reduction of T-SiO<sub>2</sub> and  $\kappa$ -Al<sub>2</sub>O<sub>3</sub> peaks. This confirmed that the synthesis of mullite in-situ growth was successful, and the in-situ reaction occurred between T-SiO<sub>2</sub> and  $\kappa$ -Al<sub>2</sub>O<sub>3</sub> exactly. With further increase in temperature, the peak intensity of mullite was obviously higher than that in 1250 °C. Temperature above 1450 °C, the peaks of T-SiO<sub>2</sub> and  $\kappa$ -Al<sub>2</sub>O<sub>3</sub> disappeared, meanwhile, AlPO<sub>4</sub> and mullite peaks became sharper, demonstrating a high in-situ mullitization and crystallization of AlPO<sub>4</sub> and mullite. Additionally, it was noticed that above 1450 °C, the mullite peak position migrated to the left with a small degree due to the Cr-doping.

### 3.2. SEM analysis

Fig. 2. shows the fracture surface morphology of the ceramics sintered from 1400 °C to 1550 °C. As seen in Fig. 2(a), a small quantity of mullite whiskers were observed at 1400 °C and their aspect ratio was short. Based on the XRD analysis, the short aspect ratio can be attributed to the incomplete in-situ reaction of mullitization, depended on a few T-SiO<sub>2</sub> residues. Moreover, it was porous and not densification, which caused poor mechanical properties of the ceramics (seen in Fig. 3). A multitude of whiskers was obtained between 1450 and 1550 °C. From Fig. 2(b), an obvious growth of mullite whisker was observed, but the aspect ratio and ceramic body was still short and porous. At 1500 °C, the length of mullite whisker grew rapidly with mean size of 5  $\mu$ m, accompanied by significant pores reduction. When temperature raise up to 1550 °C, the matrix became denser because of a continuous pores reduction, and the length of mullite whiskers were no significant changes. Following the numerous researches on mullite formation mechanism, the synthesis of mullite has been assigned as two different temperature-dependent steps [26,27]: a nucleation temperature field (<1450 °C) and a high mullitization region (1500–1550 °C). Thus, the porous and low mullitization of the ceramics below 1450 °C can be attributed to the nucleation of mullite. As described, the rapid growth of mullite whiskers above 1450 °C (as seen in Fig. 2(b)–(d)) can be attributed to the high mullitization. Furthermore, above 1450 °C, the ACP matrix became denser, indicating that the sinterability of ACP matrix was improved effectively, which is totally different from that of pure ACP [6]. Following a previous study on mullite synthesis and processing [28], this phenomenon can be presumably ascribed to the process of high

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