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# Effects of pulse conditions on microstructure and mechanical properties of $Si_3N_4$/6061Al composites prepared by spark plasma sintering (SPS)

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ABSTRACT

In this paper, 30% $Si_3N_4$/6061Al composites were prepared by SPS process to study the influence of different pulses on the microstructure and mechanical properties of the composites. The results showed that there was a sintered connection of the reinforcement $Si_3N_4$ in the composite. Moreover, with the decrease of the value of pulse condition $t_{on}$: $t_{off}$, the increased current would lead a rise of the local temperature in the composites, thus the connection of the $Si_3N_4$ particles became serious. At the same time, the interfacial reaction became serious, and the interface morphology changed from smooth to steep and then to serration. The results of mechanical properties analysis showed that different pulse conditions had little effect on the tensile strength of composites, but they had a greater effect on the elastic modulus and plasticity. When the pulse condition was $t_{on}$: $t_{off}$ = 2:1, the composite had the best mechanical properties and was superior to the performance given in many literature currently. This paper attempts to provide a certain experimental basis for the preparation of aluminum matrix composites by SPS.



## 1. Introduction

In addition to requiring higher specific stiffness, specific strength, thermal conductivity, and low density in specific structural parts of certain new aircrafts, the coefficient of thermal expansion (CET) that match the requirement ($12.5–15 \times 10^{-6}\,°C^{-1}$) and plasticity are also very important requirements. As can be designed to meet the required requirements by the type and content of reinforcements, particle reinforced aluminum matrix composites (PRAMC) are the most suitable materials for the preparation of such parts among numerous structural materials [1,2]. At present, composites satisfying the above-mentioned application requirements are mainly high volume fraction Al matrix composites, but excessively high volume fraction makes the composites brittle, and the reliability is difficult to guarantee. Therefore, it is necessary to find other suitable reinforcements to meet the needs, especially the coefficient of thermal expansion and the plasticity.

Due to its excellent properties such as high mechanical strength, good thermal and chemical stability, and low density, $Si_3N_4$ is an ideal reinforcement in particle reinforced aluminum matrix composites [3–5]. In particular, because it has a lower thermal expansion coefficient compared to other reinforcements of particle reinforced aluminum matrix composites, by adding smaller content of $Si_3N_4$, the composites can obtain the thermal expansion coefficient required for the application conditions, which will increase the plasticity and ensure the reliability of the composites. Therefore, $Si_3N_4$/Al composites have a wide range of application prospects in certain spacecrafts and other applications. Yang et al. [3] fabricated 36% $Si_3N_4$/2024Al and pointed out the thermal conductivity of annealed $Si_3N_{4p}$/2024 composite was 94.997 W/(m·K) at room temperature. Xiu et al. [4] fabricated Al matrix composites reinforced with 45% $Si_3N_4$ particles with average particle size of 1.5 μm and pointed out the tensile strength and elastic modulus of $Si_3N_{4p}$/2024Al composite was 360 MPa and 168 GPa, respectively.

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However, fracture of $Si_3N_{4p}$/2024Al composite was characterized by brittle fracture without elongation, which is unfavorable for the application of materials. Chen et al. [5] fabricated 45% $Si_3N_4$/2024Al composite and indicated that the CET of the composite was about $10.4 \times 10^{-6}\,°C^{-1}$, which was consistent with the average value of Kener model and lower bound of Schapery model. Therefore, when $Si_3N_4$ is selected as a reinforcement, the composite can satisfy performance requirements by reducing volume fraction of $Si_3N_4$.

The main manufacturing methods for particle reinforced aluminum matrix composites include stirring casting [6,7], squeeze casting [8,9], hot press sintering [10,11], hot isostatic pressing [12] and spark plasma sintering (SPS) [13–16]. Among these, SPS is a very attractive method because it can achieve rapid heating and cooling, as well as require a short holding time to make the material densified, which can prevent the growth of aluminum grains. In fact, some scholars have already adopted SPS technology to prepare PRAMC, and also have a certain researches on the mechanism of SPS process [16,17]. A more general understanding is that part of the heating power comes from the Joule heat generated by the die (which may account for a major part), and the other part comes from the Joule heating at the particle contact and the high energy spark plasma at the particle gap [18]. The plasma can destroy the oxide layer on the surface of the particles and promote the formation of sintered necks during the processing of the SPS [19]. These characteristics make SPS a more attractive alternative to traditional methods such as hot pressing, casting and extrusion. For application to aluminum matrix composites, several recent studies have shown that the SPS method allows many properties to be improved. Ehsan Ghasali et al. [20] prepared the bulk Al-SiC-TiC composites by SPS and conventional sintering. SEM investigations of SPS sintered samples showed the homogeneous distribution of reinforcement particles with neither voids nor cracks in the microstructure while conventional sintered samples were porous in some areas. The SPS compared to conventional sintering leads to good mechanical properties and fine microstructure at short sintering time. Kurita et al. [21] produced multi-walled carbon nanotube-reinforced aluminum matrix composites with an ultimate tensile strength that was 40% higher than that of pure aluminum with the conservation of remarkable fracture elongation of aluminum. Kiyoshi Mizuuchi et al. [22] prepared diamond particle dispersed aluminum matrix composites fabricated in solid-liquid coexistent state by spark plasma SPS. Thermal conductivity of the composite containing 45.5 vol.% diamond reached 403 W/mK, approximately 76% the theoretical thermal conductivity. Chen et al. [23] showed that the formation of an $Al_4C_3$ phase in CNT–Al composites prepared by SPS, even under a comparatively low sintering temperature of 500 °C. What's more, high-performance CNT-Al composites can be obtained by selecting suitable sintering conditions. In all, the generation of plasma will cause great changes in the microstructure and interfacial reaction between the materials prepared by SPS and traditional powder metallurgy, and then affect the performance of the composites.

There are many factors in the preparation of composites by SPS, and the pulse conditions are one of the most critical influencing factors. As the value of the current $t_{on}$: $t_{off}$ decreases, the intensity of the instantaneous current during sintering increases, which results in a rapid increase in the local temperature between powder particles, and even can reach a high temperature of thousands of degrees Celsius. This excessive temperature may adversely affect the performance of the composites. Gregory Lalet et al. [24] showed that the shorter $t_{on}$: $t_{off}$ times allowed larger quantities of $Al_4C_3$ crystals to be formed at the Al/Cf interface, which was detrimental to composites' properties. Therefore, it is of great significance to study the influence of pulse conditions on the properties of $Si_3N_4$/Al composites prepared by SPS.

In this paper, 30% $Si_3N_4$/6061Al composites prepared by ball milling and SPS method were present. The influence of different pulse conditions on the microstructure and mechanical properties of the composites was studied. The mechanism of the sintering of $Si_3N_4$ ceramics in composites was proposed. A composite with high strength and high elongation was prepared, whose performance was superior to many current literature. This paper attempts to provide a certain experimental basis for preparation of particle reinforced aluminum matrix composites by SPS.

## 2. Materials and methods

### 2.1. Materials

Gas-atomized 6061Al alloy powders (Northeast Light Alloy Co.,Ltd. China) and $Si_3N_4$ particles (99.9% in purity, Fujian Schnorrer New Material Co., Ltd. China) were used as the matrix and reinforcement respectively. The chemical composition (wt.%) of 6061Al alloy is shown in Table 1. 6061Al particles exhibited near spherical morphology with an average diameter of 10 μm and $Si_3N_4$ particles shaped as hexagonal short rod, as shown in Fig. 1(a) and Fig. 1(b) respectively. As shown in Fig. 1(c), the particle size distribution of $Si_3N_4$ particles is between submicron and micron, most of which distributed between 2μm and 5μm. XRD result (Fig. 1(d)) shows that $Si_3N_4$ is β-phase.

### 2.2. Fabrication process

30% $Si_3N_4$/6061Al composite ingots ($\phi$100 mm × 20 mm) used in this study were fabricated through ball milling and SPS sintering process. Meanwhile, the same volume fractions of composites were prepared using vacuum hot pressing (HP) for comparing certain phenomena that occurred in SPS. During the ball milling process, $Si_3N_4$ and 6061Al powders were mixed using a QM-3SP2 high energy ball milling machine. Ball milling was carried out in nitrogen at a rotating speed of 300 r/min for 120 min with ball-to-powder weight ratio of 10:1. $Al_2O_3$ balls, with diameters of 10 mm, 6 mm and 3 mm were used as grinding media. The SEM micrograph of mixed powder is shown in Fig. 2. It can be seen that the $Si_3N_4$ particles were distributed evenly in the 6061Al powder, and there is no obvious agglomeration and crushing phenomenon in the ball milling process. Subsequently, the mixed powders were placed in a graphite mold and prepressed in an atmospheric environment at a pressure of 5 MPa. The powder-filled mold was then placed in the SPS apparatus (FCT System Co. Ltd. (HPD 400)) and the initial pressure was also 5 MPa. When the equipment vacuum reached the required condition, it started to heat up. An infrared thermometer was used to measure the temperature of the upper pressure head of the mold and adjusted current and voltage for controlling heating and temperature during the entire SPS run. The sintering temperature was set to 570 °C, and the specific temperature curve is shown in Fig. 3(a). It should be noted that Fig. 3(a) shows the actual temperature curve. As the infrared thermometer can not measure the temperature below 400 °C, when the mold temperature is lower than 400 °C, the actual displayed temperature value is always 400 °C. As a result, in Fig. 3(a), the temperature value is maintained 400 °C from 0 to 15 min. During the heating stage, the pressure first reached 20 MPa and was maintained. When the temperature rised

**Table 1**
The chemical composition (wt.%) of 6061Al alloy.

| Chemical elements | Cu | Mg | Mn | Fe | Si | Zn | Cr | Al |
|---|---|---|---|---|---|---|---|---|
| 6061Al | 0.22 | 1.01 | 0.15 | 0.097 | 0.58 | 0.026 | 0.11 | Bal |