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In situ investigation on the mixed-interaction mechanisms in the metal–ceramic system's microwave sintering

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Abstract

In situ investigation of the microstructure evolution and the mixed-interaction mechanisms of metal–ceramic materials during the microwave sintering process was carried out using the synchrotron radiation computed tomography technique. The results indicate that there are some special mixed-interaction mechanisms, which may promote the sintering process during the microwave heating of metal–ceramic materials. In the experiment, some particular sintering phenomena that differ from the microwave sintering of pure metal materials were observed, such as fast interface bonding and particle swallowing. Quantitative analysis of the microstructure evolution during microwave sintering showed that the decrement of grain surface bending energy of metal–ceramic materials was slower, while the sintering-neck growth rate was higher than that of the pure metal materials. These results may be caused by the mixed-interaction mechanisms between microwave and metal–ceramic materials, such as the "micro-focusing effect", the special microwave interaction mechanisms on the particle surface and the heterogeneous metal–ceramic interface. This study will help to provide a useful reference for the improvement of the microstructure characteristics of metal–ceramic materials in microwave sintering.

Keywords: Microwave sintering; Metals; Ceramics; Microstructure; Synchrotron radiation computed tomography

1. Introduction

Microwave sintering is being developed as a novel method for the rapid preparation of high-performance powder materials such as metals, ceramics and metal–ceramic composite materials [1–4]. For example, Cheng et al. [5] successfully sintered transparent Al_2O_3 with a very neat particle boundary and uniform particle growth at 1750 °C for only 45 min (conventional sintering >1900 °C, several hours). Roy et al. [6] confirmed that, after 10–30 min microwave treatment, the modulus of rupture of Fe–Ni increased by 60% that of conventional specimens. Recently, Breval et al. [7] indicated that there was hardly any WC particle growth in the microwave sintering of metal–ceramic WC–Co material, and the specimen had 1–5 GPa better hardness and six times greater resistance against corrosion than the conventional specimens. Many studies have been carried out on the microwave sintering mechanisms of metals and ceramics. They indicate that the superior performances of microwave sintered materials are mainly due to the "non-thermal effects" induced by the high-frequency alternating microwave electromagnetic fields. Moreover, there are different sintering mechanisms between metals and ceramics, such as the enhancement of diffusion coefficient [8], the reduction in activation energy [9] and the "micro-focusing effect" [10] for ceramics, and the enhancement of diffusion coefficient [11] and the eddy current [12,13] for metals. However, as for the metal–ceramic materials, most of the current studies focus just on the dielectric or mechanical properties of these materials, and work seldom concerns the sintering mechanisms, especially the

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interaction mechanisms between microwave and materials in this mixed system. However, owing to the role of microwave fields, as well as the heterogeneity and non-uniform distribution of the mixed materials, there might be some special interaction mechanisms that are different from microwave sintering of pure ceramics and metals, because studies have shown that different types of materials have different microwave heating characteristics [13]. What is more, if different types of materials (such as the metals and ceramics) are mixed together, the heating characteristic will be different from or even opposite to the original materials. For example, when WC and Co were mixed together, the heating efficiency of WC/Co in a magnetic field strangely became lower than both WC and Co. For the Al_2O_3 specimen, the heating efficiency of the magnetic field was lower than the electric field at first, but as metals were added, the heating efficiency of the magnetic field became greater than the electric field [13]. These results indicate that, in the metal–ceramic mixed system, the mixed-interaction mechanisms are not just a simple superposition of the original mechanisms. There might be new mechanisms that are quite different from the original single system. These mechanisms may weaken or promote the sintering process and subsequently affect the microstructure and macro-performance of materials. In view of the above, work on the exploration of the mixed-interaction mechanisms in the microwave sintering of metal–ceramic materials is necessary.

Though microwave sintering has many attractive characteristic, in essence it is the microstructure evolution and densification process of materials driven by the coupling fields of thermal, electrical and magnetic. The direction of this microstructure evolution is closely related to the interaction mechanisms of microwave material. Therefore, in order to reveal the mixed-interaction mechanisms of the metal–ceramic materials in microwave fields, the most effective way is to observe in situ and directly the internal microstructure evolution phenomena during the microwave sintering process, extract the characteristics of the microstructure and then analyze the corresponding mix–interaction mechanisms and sintering kinetics. However, owing to the high-temperature environment and microwave radiation, it is difficult to carry out direct and in situ observation of the specimen by conventional observation techniques (e.g. scanning electron microscopy (SEM), transmission electron microscopy (TEM)). With some techniques, such as X-ray diffraction, hot-stage microscopes can carry out the in situ analysis of the surface information of materials during microwave sintering, and they have important application in the in situ research on microwave sintering. The latest non-destructive testing technology is the synchrotron radiation–computer tomography (SR-CT) technique [14]. This technique can achieve non-destructive, real-time and three-dimensional (3-D) observation of microstructure evolution under extreme conditions (e.g. high temperature, high pressure, intense radiation). Applying this technique, the internal 3-D microstructure (both the surface and the internal microstructure) evolution phenomena during the high-temperature microwave sintering process can be directly and continuously observed, and will provide rich and complete experimental data for analysis.

In the present paper, the SR-CT technique was adopted to carry out in situ investigation of the microwave sintering mechanisms of metal–ceramic materials (silicon carbide–aluminum, Al–SiC). The experiment was carried out on the BL13W1 beam line at the Shanghai Synchrotron Radiation Facility (SSRF, China, a third-generation light source with excellent features of high intensity, high brilliance, high polarization rate and quasi-coherence). In the present experiment, two-dimensional (2-D) and 3-D images of the microstructure of Al–SiC at different sintering times were reconstructed. From these images, some special sintering phenomena of the mixed system were clearly observed, such as fast interface bonding and particle swallowing. Moreover, the microstructure characteristics (e.g. surface area, surface curvature) of the specimen were extracted, and the quantitative analysis of the changes in particle surface bending energy and sintering-neck growth kinetic mechanisms of the mixed system was carried out and compared with the single material system of metal. These results will help to reveal the mixed-interaction mechanisms of microwave sintering of metal–ceramic materials and offer some useful references for actual production.

2. Experimental and results

The experiment on the microwave sintering of Al–SiC specimen was carried out on the BL13W1 beam line at SSRF (energy 28 keV). Chemically pure SiC (purity 99.9%, average diameter 120 μm) and Al (purity 96%, average diameter 120 μm) were used. Before the experiment, Al–SiC (volume ratio of 1:1, mixed uniformly) powders were loosely poured into a closed capillary (internal radius 0.5 mm, height 10 mm) and then introduced into a specially designed microwave furnace (multimode cavity 2.45 GHz, output power 0–3 kW) [15]. The SR–CT experiment system is shown in Fig. 1. The experimental process and the power profile of the microwave are the same as in the experiment on Al, which was carried out previously [16]. In the present experiment, the temperature was measured by a thermo tracer (type TH5104, temperature measurement range −10–1500 °C, accuracy ±1.0% (full scale), emissivity 0.30 for Al and 0.51 for Al–SiC), and the typical temperature profile is shown in Fig. 2.

One thing that needs explanation is that, in the present experiment, the powders were loosely placed in a two-end-closed quartz capillary (internal diameter 1.0 mm, height 10 mm). According to the calculation, there was only $\sim 7.5 \times 10^{-7}$ l oxygen in this closed quartz capillary (the powders can be considered as body-centered cubic (bcc) packing, as shown in Fig. 3a), so the oxidation of Al would stop as the very small amount of oxygen was consumed ($\sim 1/10^4$ of Al was oxidized, or only ~2 nm thickness of the 120 μm Al particles was oxidized). So the value of normalized thickness of the insulating shells *d*