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Influence of Transferred Arc Plasma Melting Time on the Formation of Phase and Microstructure of Mullite-Zirconia Composite

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Abstract: Transferred arc plasma is an effective and simple technique to synthesis a high temperature reaction ceramic composite material. In this paper, 20 kW transferred arc plasma torch was used to synthesis mullite-zirconia composites through the solid-state reaction of 3:2 mole ratio of ball milled alumina and zircon powders. Dissociation of zircon in a thermal plasma arc is utilized as to prepare mullite-zirconia composites. The ball milled samples are melted for 3, 6, 9 and 12 minutes in transferred arc plasma torch at 20 kW power level with 10 lpm of argon flow rate and cooled by air. The phase and microstructure of melted samples were determined from X-ray diffraction (XRD) and SEM images. The obtained results shows that the processing time significantly influence on the formation of phase and microstructure of the mullite- zirconia composite.

Keywords: Mullite- Zirconia, Transferred Arc Plasma, Plasma Melting.

Introduction

Mullite ($3Al_2O_3$. $2SiO_2$) is the only stable compound in the alumina–silica system at room temperature and pressure. Mullite ceramics possess excellent creep resistance and chemical stability and are suitable for high temperature applications [1-5]. However, the low fracture toughness of mullite and, the difficulties in sintering to full density are the main obstacles for mullite materials for more widespread engineering applications. ZrO_2 addition is effective in improving the strength and fracture toughness at intermediate temperatures. Various processing routes [6-9] can be used to prepare mullite–zirconia composites. Reaction sintering of zircon and alumina is a relatively simple and inexpensive route to obtain homogeneous mullite-zirconia composite with enhanced mechanical properties. However, it's required very high temperature, so we need to develop a high temperature medium with controllable parameters. Regarding the above purpose transferred arc plasma (TAP) torch is a one of the simple and cost-effective tools for high temperature ceramic reactions. In the present investigation, methods for synthesis of mullite-zirconia composite based on transferred arc plasma melting of alumina and zircon mixtures has been proposed and the influence of melting time was studied.

Experimental set up and procedures

In this experiment 20 kW transferred arc plasma torch (Ion Arc Technologies Pvt. Ltd., India) used for melting purpose. The schematic diagram of the melting torch is shown in Figure.1. The torch consists cylindrical well in a graphite lot, which is the anode (100 mm height and 70 mm outer diameter). The cathode is made of graphite rod 250 mm long and 50 mm diameter. Its end tapers to a conical shape for better electron emission. The cathode is enclosed in a hollow brass cylinder and provisions are made for water circulating (for cooling) and gas flow. The system has multiple inlets for plasma gas at the cathode end.

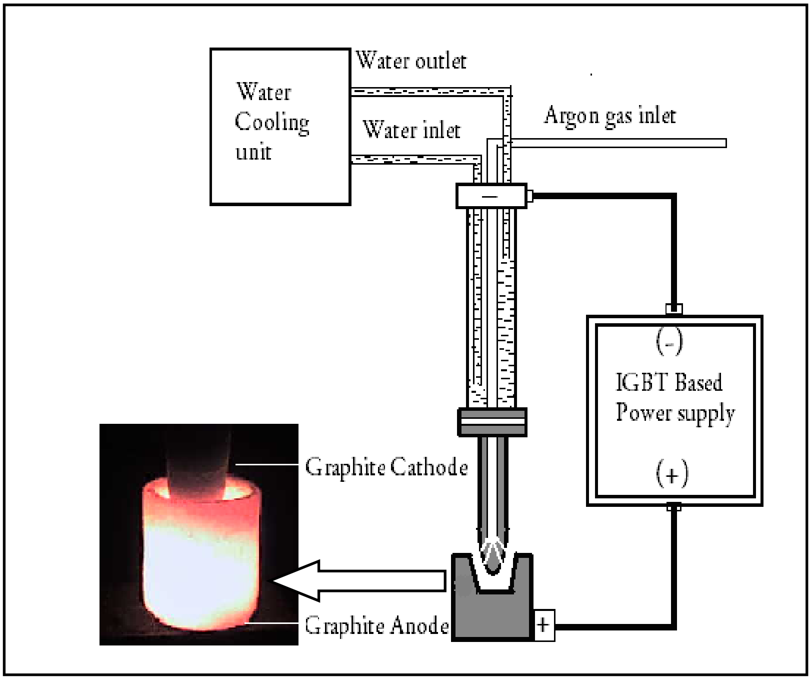

Figure 1. Schematic diagram of the transferred arc plasma melting torch.

The materials used in this study were relatively pure, commercial grade zircon sand (>98 % $ZrSiO_4$, Ion Arc Technologies pvt.ltd, India) and alumina (99% Al_2O_3 Loba Chemical Pvt. India). The average particle sizes of the raw $ZrSiO_4$ and Al_2O_3 powders were 75-100 µm and 30-

50 µm respectively. The Al_2O_3- $ZrSiO_4$ powders with 3:2 molar ratio was milled with Corundum ball milling media using the Planetary Mill (Insmart, India). The powder to ball ratio was kept to 1:10 by weight. The bowl was put on the mill and rotated at the speed of 200 rpm for 4 h. The composition of the sample for alumina-zirconia composite is given by the following reaction:

$$2ZrSiO_4 + 3Al_2O_3 \rightarrow 2ZrO_2 + 3Al_2O_3.\ 2SiO_2$$

The milled powder was then melted in 20 kW transferred arc plasma torch (TAP) at 3, 6, 9 and 12 minutes with 10 lpm argon gas flow rate by open atmosphere. Then the melted samples were cooled by open air. X-ray diffraction (XRD) analysis of the melted samples was performed using an PW Philips type diffractometer with Cu-Kα radiation. The microstructures of the melted samples are studied by the Philips XL40 type scanning electron microscope (SEM).

Results and discussions

Figure. 2 (a-d) shows the XRD pattern of transferred arc plasma melted mullite-zirconia composites with different melting time. Interestingly, in addition to the mullite peaks, a monoclinic phase zirconia and zircon peaks are present in 3-minute melted samples. Appearance of zircon peaks may due to the insufficient temperature of plasma arc reactor for complete dissociation. Furthermore, traceable amount of quartz peaks appears along with mullite and monoclinic zirconia peaks in 6-minute melted sample (Figure. 2. b). Meanwhile, the expected mullite and the monoclinic zirconia phase appears in 9 and 12 minutes of transferred arc melted samples. Observed results clearly reveal that the short melting time of transferred arc plasma torch is not enough for complete mullite-zirconia composite formations. Moreover, the tetragonal phase zirconia did not appear in any samples which treated in transferred arc plasma due to the presence of SiO_2 in the reaction system [10, 11].

Figure. 3 (a- d) shows the SEM images of transferred arc plasma melted mullite-zirconia composite with different melting time. The mullite-zirconia composites appear in different microstructure with respect to melting time. The star shaped zirconia and needle shaped mullite whiskers appear in samples melted for 3- minute (Figure 3. a). Meantime, the sample meted at 6-minute consist of flower shaped zirconia embedded in mullite needles occur. (Figure. 3. b). Further increasing the melting time from 6 to 9 and 12 minutes, flower shaped zirconia transform to a square shaped structure with uniform array of lines (Figure. 3 c & d). This results clearly shows that the melting time strongly influence the formation of microstructure in mullite-zirconia composites. The EDS spectrum of the melted samples (Figure.4) confirmed that the arrays of white lines are zirconia and the dark colored needles are mullite.

In general, ZrO_2 and amorphous SiO_2 were formed due to $ZrSiO_4$ dissociation, and ZrO_2 particles were immersed in the amorphous SiO_2 matrix, which SiO_2 stabilized ZrO_2 as t-ZrO_2 phase. Meantime, at high temperatures, the amorphous SiO_2 was finally consumed to form mullite, losing the ability to stabilize ZrO_2. Hence, the ZrO_2 which obtained at high temperatures thermal plasma melting process preferred to be monoclinic phase (m-ZrO_2).

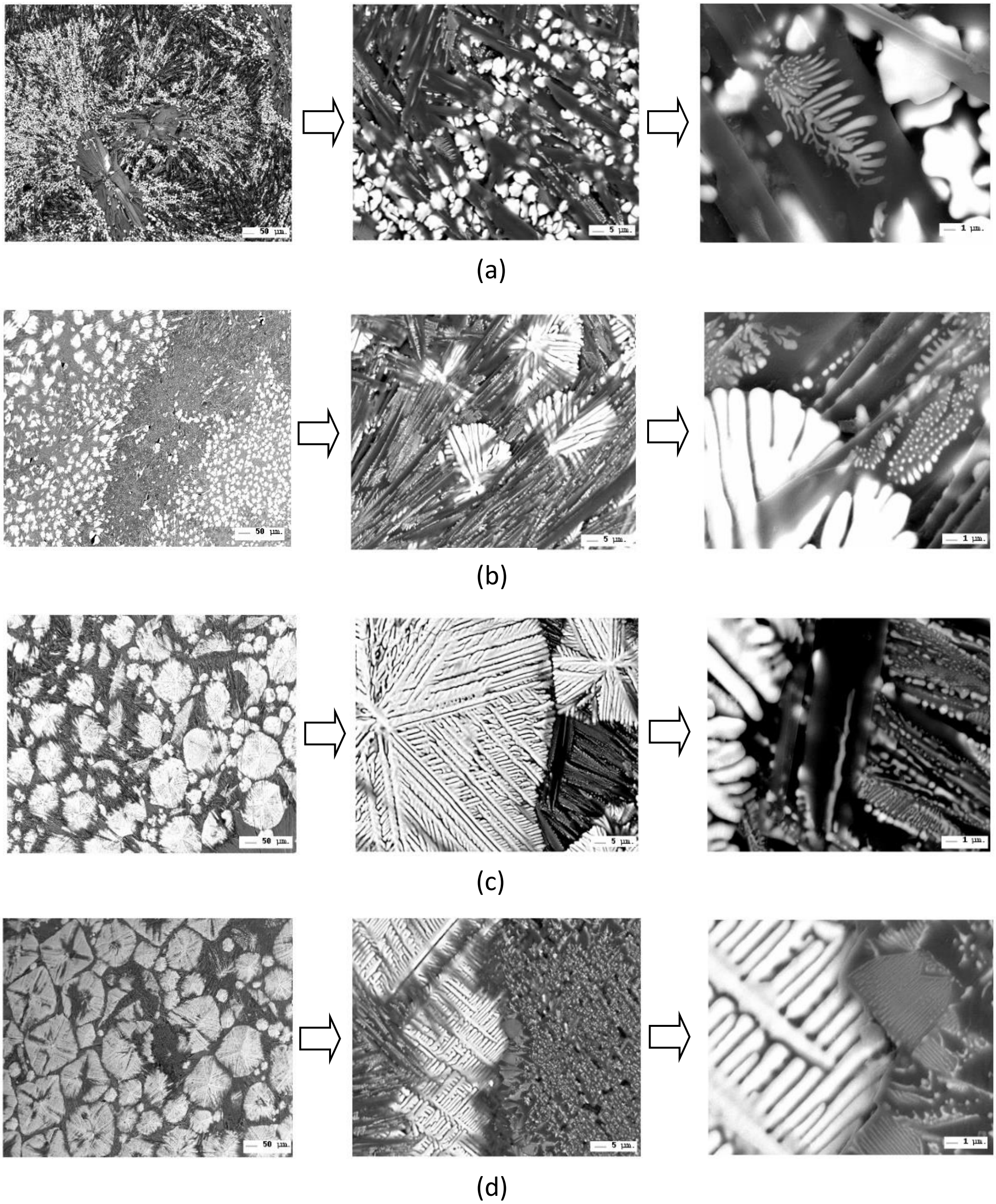

Figure 3 Microstructure and corresponding enlarged view of transferred arc plasma melted mullite-zirconia composite, a) 3 minutes, b) 6 minutes, c) 9 minutes, d) 12 minutes

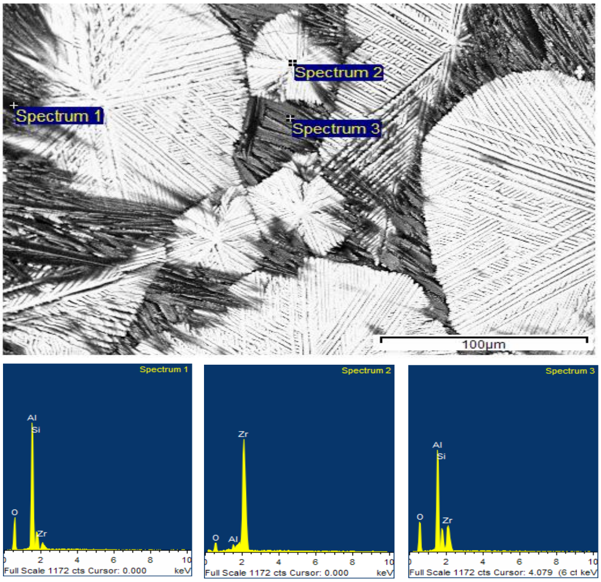

Figure 4. EDS Spectrum of the transferred arc plasma melted mullite-zirconia composite

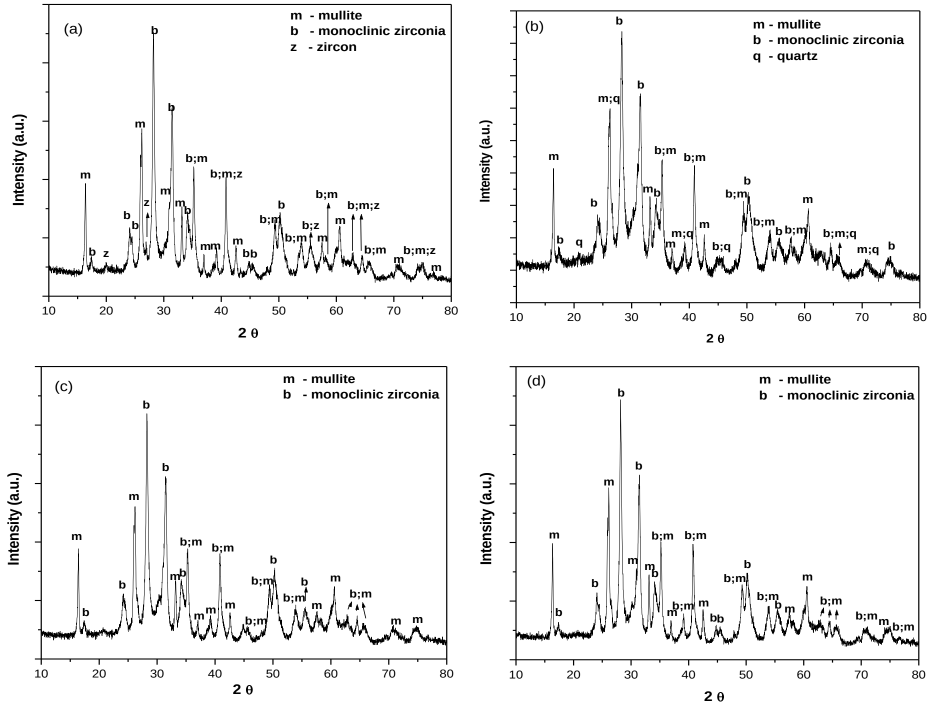

Figure 2 XRD pattern of transferred arc plasma melted mullite-zirconia composite a) 3 minutes, b) 6 minutes, c) 9 minutes, d) 12 minutes

In addition to that, ZrO_2 phase transformation is subjected to a particle size effect; a larger particle size enhances the t-ZrO_2 → m-ZrO_2 transformation on cooling. Herein, the high temperature thermal plasma reaction followed by normal cooling, ZrO_2 grains would grow uniformly larger with increase in melting time, and larger ZrO_2 grains easily converted to m-ZrO_2 phase when cooled down to room temperature. These considerations can reasonably explain the phase change of ZrO_2 during the reaction although the exact reasons are not very clear. Mullite formation has been described by interdiffusion of aluminum and silicon ionic species or nucleation and growth mechanism within the alumina/silica interfaces. According to the above stated mechanisms of mullite formation, in the present system might follow the interdiffusion of aluminum and silicon ionic species because of high temperature of transferred arc plasma. During the melting, the zircon dissociation began at melting and formed amorphous SiO_2. The amorphous SiO_2 formed softened with increasing temperature, then penetrated into the Al_2O_3 agglomerates and dissolved the Al_2O_3 to form an amorphous aluminosilicate phase. Mullite nucleating will occur when the concentration of alumina in the amorphous phase exceeds the critical nucleation concentration (CNC) or when the liquid structure becomes saturated with mullite.

Initial formation of glassy phase from the present impurities with partially thermal dissociated zircon

↓

Dissolution of zircon into this glassy phase

↓

Dissociation of zircon and the formation of ZrO_2 and higher SiO_2 – rich glassy phases

↓

Dissolution of Al_2O_3 in this glassy phase

↓

Concentration of Al_2O_3 in the melt increases until the stoichiometric composition of mullite is obtained

Mullite formation is more complex in the reaction sintering of Al_2O_3-$ZrSiO_4$ mixtures because it involves a solid-state dissociation of zircon compared to those Al_2O_3–SiO_2 reaction systems. The processes involved on the formation and growth of mullite grain can be expressed in following flow pattern. Since the understanding of actual rection mechanism of Al_2O_3- $ZrSiO_4$ system under high temperature process involves high degree of complexity, the proposed mechanism of mullite growth here is only a reasonable hypothesis and needs to be confirmed by further

Much attention has been always given to the formation of mullite in the Al_2O_3 - SiO_2 binary system because of its great importance, and several mechanisms of mullite formation have been proposed [12-14] However, probably due to the complexity of Al_2O_3- $ZrSiO_4$ system, in which zircon, alumina, zirconia and pores are simultaneously present during reactions, the solid-state reaction concerning the formation of mullite has not been intensively documented. Mullite formation is more complex in the reaction sintering of $ZrSiO_4$- Al_2O_3 mixtures because it involves a solid-state dissociation of zircon compared to those simple Al_2O_3–SiO_2 reaction systems. In ZrO_2–SiO_2 phase diagrams [15, 16], it was shown that $ZrSiO_4$ dissociation occurred above 1600ºC.

Conclusion

Mullite-zirconia composite was synthesized by 20 kW transferred arc plasma torch at different melting time. The processed mullite-zirconia composites phase was identified by XRD and the structural morphology can be identified by SEM images. The results show that increasing the melting time strongly affects the microstructure and reduce the zircon percentage. The uniform alignment of zirconia, mullite grains increases with increasing the melting time. Further research is needed to correlate the operating parameters and microstructure formation of mullite and mullite-zirconia composites.

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