Mullite Glass Ceramics Production From Zircon Sand And Alumina By High Temperature Plasma

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Abstract- *Mullite was prepared by transferred arc plasma (TAP) torch from natural alumina and zircon sand. The mixture was based on 3:2 mole ratios of alumina and zircon were ball milled and melted for 3 min in a transferred arc plasma torch. The obtained samples were investigated by XRD and SEM respective to analysis the phase and microstructure formation and the crystalline behavior of the sample was evaluated by differential thermal analysis. The obtained results clearly showed that the complete mullization was achieved alumina–zircon system.*

Keywords- Mullite, Zircon, Transferred arc plasma, X-ray diffraction

I. INTRODUCTION

Mullite ceramics have been extensively studied because of their excellent properties of high melting point (1830 °C), moderate thermal expansion coefficient, high resistance to thermal shock, good chemical durability, low thermal conductivity, low dielectric constant, excellent creep resistance and sufficient mechanical strength [1]. 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 of mullite at intermediate temperatures [2]. Various processing routes can be used to prepare mullite–zirconia composite [3-6]. The conventional reaction sintering methods take a long time to produce the composites. Alternately, Transferred arc plasma (TAP) processing is an effective and simple technique to synthesize a mullite-zirconia composite at short processing time within low power level [7, 8]. The understanding of the microstructure-property and microstructure-processing parameters relationship is essential for the improvement of ceramic materials for structural applications [9, 10]. Composite materials with a microstructure designed to give mechanical properties suited to the performance requirements could be synthesized. Optimization of processing parameters is playing a major role to control the microstructure and properties of the materials. In TAP processing, an input power of the plasma torch and processing time period are the main parameters on the preparation of mullite-zirconia composite.

The Various processing routes [11–13] can be used to prepare mullite–zirconia composites. Reaction sintering of zircon and alumina is a relatively easy and inexpensive route to obtain homogeneous mullite–zirconia ceramics with enhanced mechanical properties. Yet, when conventional reaction sintering methods are employed, problems are encountered in the form of low bulk and grain boundary diffusion coefficient of mullite, and high processing temperatures (up to1500 °C). As a result, sintering additives such as CaO, MgO, TiO2 and CeO, SrO are utilized to develop a transient liquid phase to produce a tough mullite–zirconia composite with high density [14,15]. The most recent spark plasma sintering (SPS) method is capable of sintering ceramic powders rapidly to its full density at a relatively lower temperature compared to the conventional sintering methods [16–18]. Transferred arc plasma (TAP) melting is a new sintering process to overcome the aforementioned problems and sintering ceramic powders quickly. The time duration and the cost-effectiveness are lower compared to the conventional sintering methods.

In this study, a method for fabricating mullite based on transferred arc plasma melting of alumina with zircon mixtures has been proposed. X-ray diffraction (XRD) analysis of the TAP melted samples was performed using a Rigaku, Dmax 2200 type diffractometer with Cu-Kα radiation. The microstructures of the TAP melted samples were studied by JEOL JSM-5310 scanning electron microscope (SEM).

II. EXPERIMENTAL

2.1. Materials and method

The relatively pure commercial grade zircon sand ($ZrSiO_4$; IREL, India) and alumina (Al_2O_3; Loba Chemie Pvt. India) were used as raw materials to prepare mullite-zirconia composite through TAP torch The Al_2O_3 - $ZrSiO_4$ powders with 3:2 molar ratio were 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 ratio of the starting materials was selected from the basis of the following reaction mechanism:

$$2ZrSiO_4 + 3Al_2O_3 \rightarrow 2ZrO_2 + 3Al_2O_3 . 2SiO_2 \text{ (mullite)}$$

Table 1 Typical operating parameters

Input power	: 5 kW
Plasma Gas and flow rate	: Argon; 10 lpm
Cooling water flow rate	: 10 lpm
Processing time	: 2,4,6 & 8 minutes
Cooling time	: 10 minutes
Cooling medium	: Air

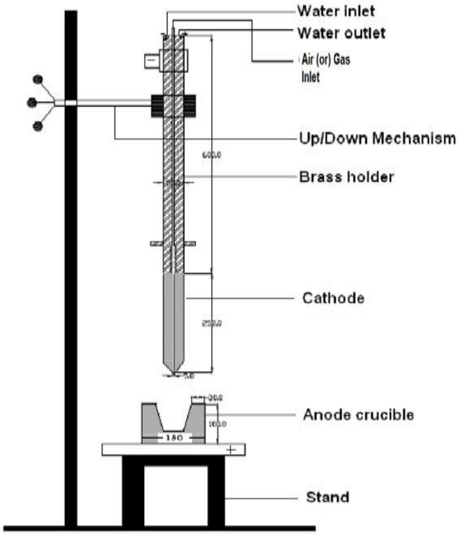


Figure 1. Schematic diagram of experimental setup.

The milled powder was then melted in TAP torch (Figure 1) at selected torch 5 kW with different processing time period (2, 4, 6 and 8 minutes). The typical TAP operating parameters are given in Table 1. The torch was operated in open atmosphere and argon gas was used as a plasma forming gas. After the process, the melted samples were cooled by applying forced air.

III. RESULTS AND DISCUSSION

3.1 XRD Analysis

Figure 2 (a-d) shows the XRD pattern of 5 kW TAP processed mullite-zirconia composites for selected processing time.

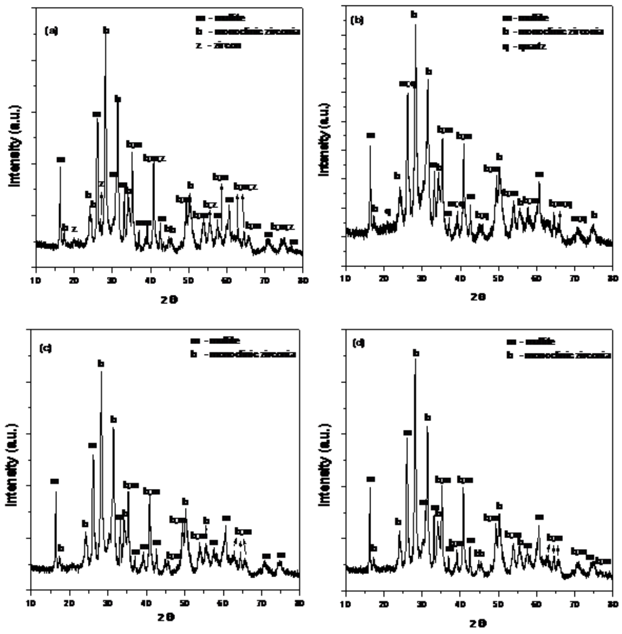
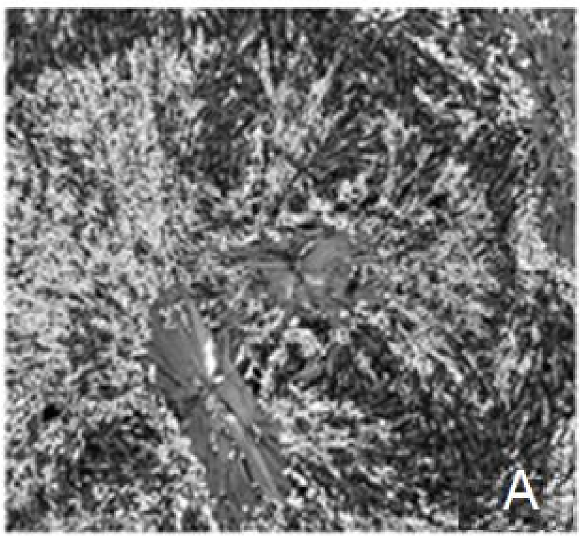
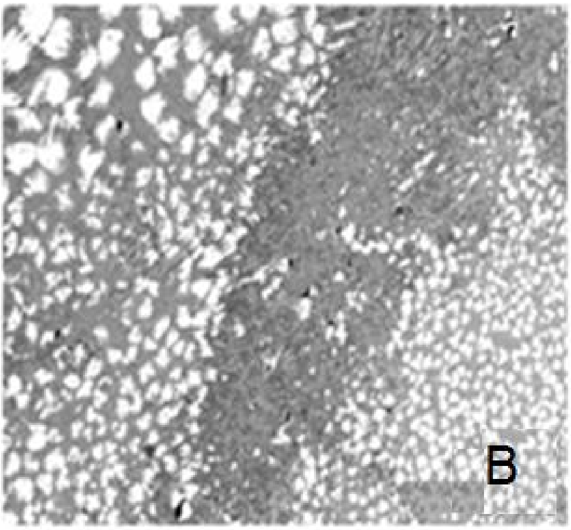
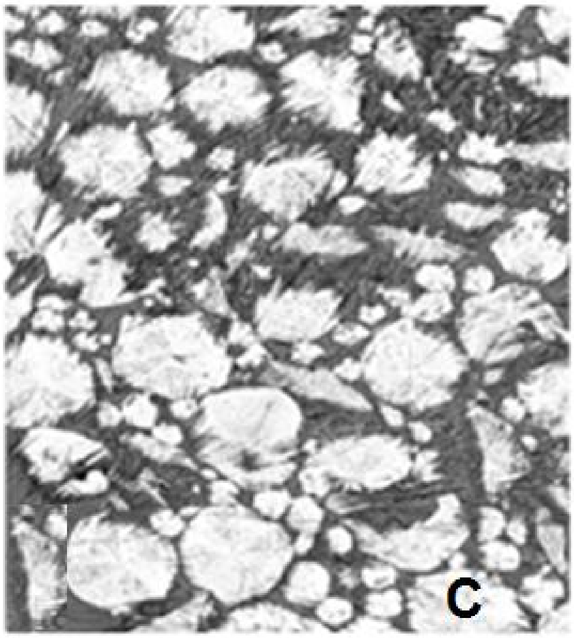


Figure 2 XRD pattern of TAP processed mullite at different processing times: (a) 2minutes; (b) 4 minutes; (c) 6 minutes; (d) 8 minutes

The pattern of 2 minutes processed sample appears with predominant monoclinic phase zirconia peaks along with mullite peaks; in addition, a small number of zircon peaks is present due to the insufficient processing temperature of plasma arc for dissociation of zircon into zirconia and silica. Similarly in 4 minutes TAP processed sample, small number of quartz peaks appear along with mullite and monoclinic phase zirconia peaks. Hence the increasing process time has completed the zircon dissociation to produce zirconia and silica.







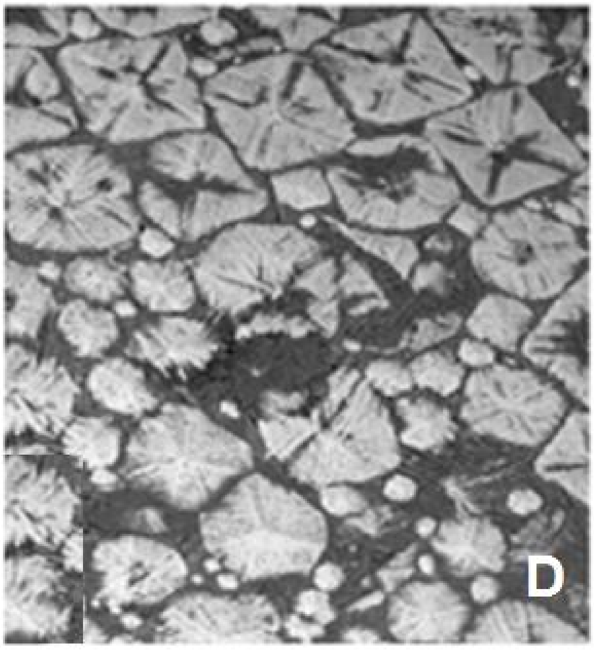


Figure 3 SEM image of TAP processed mullite at different processing times: (a) 2minutes; (b) 4 minutes; (c) 6 minutes; (d) 8 minutes

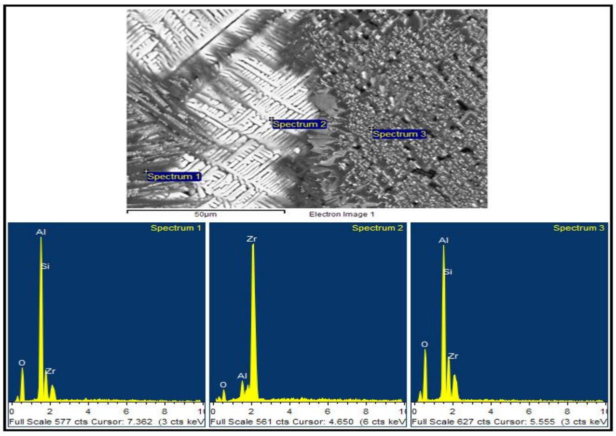


Figure 4 EDX spectrum of 5 minutes TAP processed mullite at 5 kW

The XRD pattern of 6 minutes TAP processed sample appears only with monoclinic phase zirconia and mullite peaks. The absence of zircon and quartz peaks confirms that the processing time is enough for complete solid state reaction between zircon and alumina during the process. The similar pattern is seen in 8 minutes TAP processed sample also. In all the samples tetragonal phase zirconia did not appear due to the large presence of SiO_2 in the ZrO_2–SiO_2 system.

3.2 SEM Analysis

Figure 3 (a- d) shows the SEM images of 5 kW TAP processed mullite-zirconia composite for selected processing time. The mullite-zirconia composites appear in different microstructure with respect to processing time. Initially, the star shaped zirconia and needle shaped mullite whiskers appear in 2 minutes processed sample (Figure 3a). Generally mullite grains formed are mostly equiaxed in nature, however the stucture strongly depend on the Al_2O_3/SiO_2 ratio. The structural arrangement of mullite and zirconia was confirmed by SEM with EDX spectrum (Figure 4). The EDX spectrum shows that the mullite appears in gray color with needle structure and the zirconia appears in white color with star and flower structure. On increasing the processing time from 2 to 4 minutes, the star shaped zirconia changed into flower shape and embedded in needle structured mullite whiskers (Figure 3b). Increasing the process time from 4 to 6 minutes, the size of flower shaped zirconia was increased and the presence of mullite whiskers also decreased. The internal arrangement of zirconia flowers also slightly changed (Figure 3c). The increasing process time enhances the evaporation of SiO_2 than Al_2O_3 and ZrO_2. Hence the evaporation of SiO_2 and Al_2O_3 is the main cause for the change in internal structure of flower shaped zirconia. Further increasing the process time from 6 to 8 minutes, the flower shaped zirconia was completely transformed to an approximate square shaped structure in uniform array of lines (Figure 3d) which is due to the continuous grain growth of zirconia and consequent decrease of mullite ratio through the continuous evaporation of SiO_2 and Al_2O_3. Hence the above results clearly demonstrate that the processing time period of TAP strongly influences the microstructure formation of the mullite-zirconia composites.

IV. CONCLUSION

Mullite-zirconia composite was successfully synthesized by TAP processing with selected input powers of the torch and processing times. The effect of torch input power and processing time on the formation of mullite-zirconia composite were studied. The results showed that the dense and complete mullization was achieved in high processing time due to the sufficient input power and processing time of the transferred arc plasma torch. But in low processing time due to inadequate input power the complete mullization was not achieved. The phase and microstructure formation of the processed samples were examined by XRD and SEM (including EDX) respectively. From the results, the optimum parameters for obtaining a mullite-zirconia composite was found to 5 kW torch power with more than 6 minutes processing. The results showed that the input power of the torch strongly influences the phase and microstructure of the mullite- zirconia composite.

REFERENCES

[1] S. Ananthakumar et.al, Acta Mater. 54 (2006) 2965.
[2] N. Claussen and J. Jahn, J. Am.Ceram. Soc. 63 (34) (1980) 228.
[3] S. Maitra et.al,Ceramics Intern. 32 (2006) 201.
[4] L.B. Garrido et.al, Materials Sci. and Eng. A. 419 (2006) 290.
[5] H.C. Park et.al, Materials Sci. and Eng. A 405 (2005) 233.
[6] F. Temoche et.al, Ceramics Intern. 31 (2005) 917.
[7] W.C. Butterman and W.R. Foster, Am. Mineral. 52 (1953) 880.
[8] R. Mueller et.al, J. Am. Ceram. Soc. 87 (2) (2004) 197.
[9] Claussen N, Jahn J. mechanical properties of sintered in-situ reacted mullite–zirconia composites. J Am Ceram Soc 1980;63 (34):228–9.
[10] Boch P, Chartier T. Tape casting and properties of mullite and zirconia–mullite ceramics. J Am Ceram Soc 1991;74(10):2448–52.
[11] Khor KA, Li Y. Effects of mechanical alloying on the reaction sintering of ZrSiO4 and Al2O3. Mater Sci Eng A 1998;256:271–9.
[12] Moya JS, Osendi MI. Effect of ZrO2 (ss) in mullite on the sintering and mechanical properties of mullite/ZrO2 composites. J Mater Sci Lett 1983;2:599–601.
[13] Di Rupo E, Anseau MR, Brook RJ. Reaction sintering:correlation between densification and reaction. J Mater Sci1979;14:2924–8.
[14] Pena P, Miranzo P, Moya JS, De Aza S. Multicomponent toughened ceramic materials obtained by reaction sintering, Part 1 ZrO2–Al2O3–SiO2–CaO system. J Mater Sci 1985;20:2011–22.
[15] Kinikoglu S, Marsoglu M. Effect of SrO on densification and mechanicalproperties of reaction-sintered mullite–zirconia. J Eur Ceram Soc 1994;14:45–51.
[16] Johnson DL. Microwave and plasma sintering of ceramics. Ceram Int 1991;17(5):295-300.
[17] Risbud SH, Shan CH. Fast consolidation of ceramic powders. Mater Sci Eng A 1995;204:146–51.
[18] Su H, Johnson DL. Sintering of alumina in microwave-induced oxygen plasma. J Am Ceram Soc 1996;79(12):3199–201.