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The Effect of Carbon Infiltration Through CVI Techniques on The Physical Properties of ZTA

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ABSTRACT

The preference for a ceramic matrix composite, in many applications, is due to its allowance for alterations, with regards to the filler or reinforcement used. Among the well-known ceramic matrix composites, is the carbon nanotube reinforced ceramic matrix composite. This ceramic composite, which possesses excellent properties, is used exclusively for high-end applications. However, other than the fact that this composite is prone to damage, in a high temperature environment during the sintering process, its use also comes at a high cost. Among the solutions to these drawbacks, is the introduction of carbon into the ceramic composite, through the chemical vapour infiltration (CVI) process. Initially, pyrolysis of biomass is applied on the empty fruit bunch (EFB), for the generation of tar vapour. Pyrolytic carbon is then produced through secondary reaction, during contact with the ceramic surface. The findings, derived from previous studies, indicate that the physical properties of ZTA improve with the infiltration of carbon, until optimum holding time is arrived at. The optimum holding time, of 2.5 hours for the CVI process, is due to the lowest surface area (3.605 m^2/g) and the highest density (3.787 g/cm^3). This can be attributed to the reduction in porosity, as the holding time increased. Thus, it can be concluded that carbon infiltration will significantly reduce the porosity, and improve the ceramic properties, of zirconia-toughened alumina (ZTA).

Keywords:
ZTA, Alumina, Pyrolitic Carbon, CVI,
EFB, porosity

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1. Introduction

Aluminium oxide, commonly known as alumina, is a tough ceramic that is well-suited for wear-resistant products and applications. The wide usage, of alumina-based ceramic composites, can be attributed to their attractive properties [1]. Alumina also belongs in an oxide ceramics group, which is recognised for its good physical and mechanical properties [2,3]. As reported by Galusek and Ghillanyova [4], alumina comes with an extremely high melting point of approximately 2053˚C. While this renders alumina appropriate for high-temperature applications, Al_2O_3 has a low fracture

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toughness of roughly 4–5 MPa $m^{1/2}$. Many investigations have been conducted on the use of zirconia as a ceramic composite, to overcome the low fracture toughness problem of alumina. This research is aimed at improving the mechanical properties of the system, with the emphasis on its strengthening processes [5,6].

The fracture toughness of zirconium dioxide (ZrO_2), which ranges from 6 to 10 $MPam^{1/2}$, is considerably superior to that of Al_2O_3 ceramic. Thus, the introduction of zirconia into alumina, will serve to enhance its mechanical properties. The toughening mechanism is activated through the transformation of zirconia, brought about by compressive stress on alumina [7]. This is attributable to the conversion of metastable tetragonal ZrO_2, into a stable thermodynamic monoclinic, which acts on the crack end area encountering stress. The expansion in volume, which comes with this conversion, serves to prevent crack propagation, and increase the fracture toughness of Al_2O_3 [8,9]. The addition of materials to zirconia ceramic has a beneficial effect on its microstructure and properties. Moreover, as reported by Nogiwa-Valdez et al. [10], the inclusion of more than one stabiliser to zirconia, delivers a Zirconia-based ceramics with optimised mechanical parameters.

The use of carbon as a filler or reinforcement in ceramic matrix composites for a variety of applications, is well established [11, 12]. The presence of carbon in composites serves to enhance the properties of the base material. This includes the reduction of the brittleness of the ceramic, as well as an improvement in the quality of materials produced. Carbon is obtainable from EFB, as this form of biomass is made up of carbon, hydrogen, and oxygen, which can be thermochemically converted into gas, energy, chemicals, and fuel [13,14,15]. Due to its high carbon content, EFB is an excellent choice for carbon infiltration in ceramic composites. Furthermore, as EFB biomass is readily available in the oil palm industry, this circumstance not only serve to reduce costs, but also mitigate environmental issues, such as the indiscriminate disposal of waste [16,17]. The infiltration of carbon into the composite is achievable through several methods. Among them is the use of hot pressing, which requires a high sintering temperature to attain a sufficient density. However, this method tends to cause damage to the carbon [18,19,20]. Carbon substitution using spark plasma, calls for the use of high energy, and involves high production costs [21].

In this study, chemical vapour infiltration (CVI) method was employed to facilitate carbon penetration within the pores of ZTA ceramic composites by tar decomposition into solid carbon. EFB was heated at temperatures between 400 to 700 ℃ to produce tar to be infiltrated and decomposed into carbon within ZTA pores [22]. The physical and mechanical properties of the carbon infused ZTA were examined and the effects of holding time of the CVI process, on the amount of carbon infiltration into the ZTA composite were investigated.

2. Methodology

Alumina with 99.8 wt % purity and yttria stabilized zirconia with 94.5 wt % purity was used in this study. Both materials were wet mixed in the ratio of 4:1 using ABB Mixer Mill. The ratio between the mixed materials and zirconia ball were 20:1, with the speed of 200 rpm for 24 hours. Subsequently, the slurry was dried in oven at 105 ^oC for 24 hours. Subsequently the dried slurry was crushed and passed through a 75 µm size sieve. The powder was then compacted into pellets using hydraulic press at 300 MPa with a thickness of 8mm and 16mm diameter. The pellets were the sintered at 1600 ^oC for 1-hour soaking time with 5°C/minute of heating rate.

The sintered pellets were subjected to the chemical vapour infiltration process for carbon penetration. EFB was first dried in an oven at 100 ^oC for a period of 24 hours. 100 grams of EFB was introduced into furnace with the previously sintered ZTA pellets at 500 ^oC in an inert atmosphere. During the heating process, EFB was pyrolyzed into tar vapor before being infiltrated within the ZTA

pore networks. This CVI technique was performed with holding times of 0h, 1h, 1.5h, 2h, 2.5h, and 3h.

Phase identification was done using X-ray Diffraction (XRD) technique (Bruker D8 Advance) with CuKα radiation= 1.5418 Å operating at 40kV, 30mA the range of 20-80° of 2θ. The presence of carbon material in the composite was examined by Raman spectroscopy. Brunauer-Emmett-Teller (BET) was done to analyse the surface area and determines the specific pore volume. The morphology on the microstructure of the samples was observed using scanning electron microscope (SEM) model InTouchScope JSM-IT100. Each sample is required to be coated with conductive material palladium (Pd). Bulk density of the pellets was measured using Densitometer. Fig. 1 shows the summary of the sample preparation and its respective characterizations.

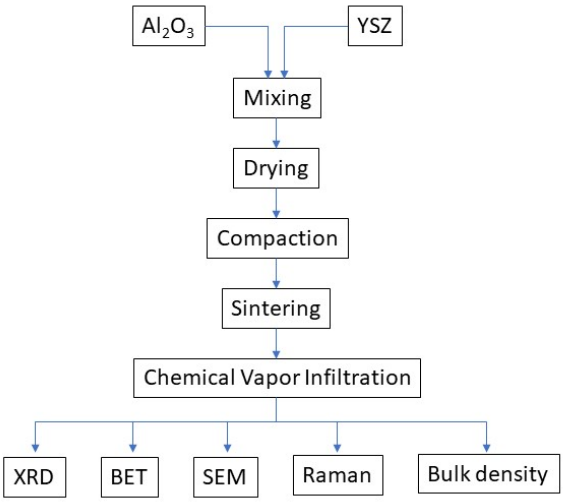

Fig. 1. Flowchart of the sample preparation and respective characterizations.

3. Results

3.1 Phase Analysis

Fig. 2 shows the XRD patterns of carbon infiltrated ZTA ceramic composites, prepared using the CVI method, under various holding times. The effect of carbon infiltration inside the ZTA can be observed from the peak intensity at a two-hour holding time. In comparison to the other holding times, the intensity at 2 hours of holding time has a lower peak. This finding is in accordance to that of a previous study conducted by Krishnan *et al*. [23], where the intensity of XRD peaks was observed to dip, as more carbon was added to the alumina. This could be due to the high carbon content infiltration into the ZTA, which restricted the sample's surface, causing the 2-hour holding time peak, to be lower than those under other holding times.

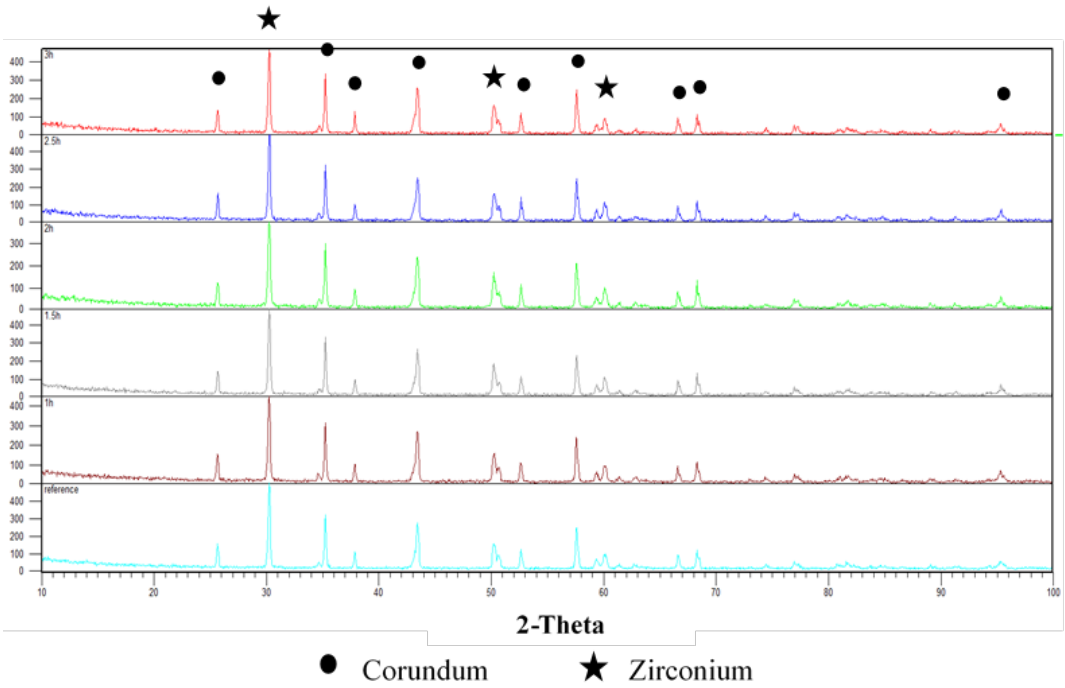

Fig. 2. X-ray diffraction analyses for ZTA-C samples with different holding time of CVI method

The effect of holding time on the c/a ratio during the CVI process is depicted in Fig. 3. As can be observed, the corundum, or alumina lattice, increased up to a maximum of 2 hours holding time. An extended holding time (more than 2 hours) resulted in a slight decrement in the corundum or alumina lattice. This finding agrees with those from previous studies, in which a holding time of more than 2 hours, led to a decline in the properties of the corundum.

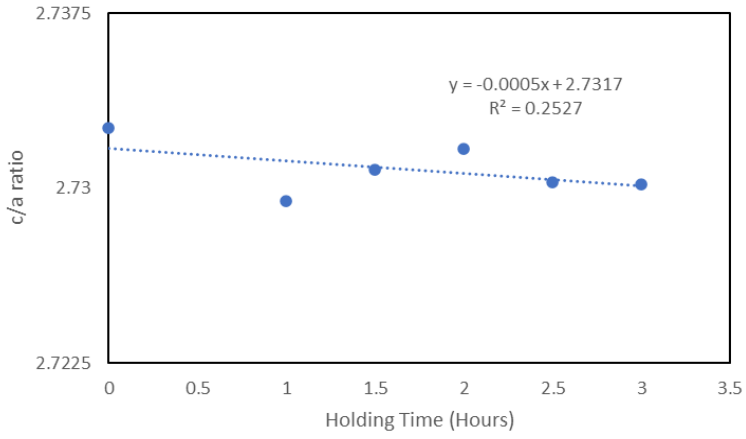

Fig. 3. c/a ratio as function at different holding time of CVI

While XRD analysis was utilized to observe the effect of carbon infiltration into the ZTA composite, Raman analysis was performed to confirm the existence of carbon in the composite. The appearance of the D band (~1330 cm^{-1}) and the G band (~1520 cm^{-1}) during the Raman analysis, as shown in Fig. 4, verifies the presence of carbon in the composite [24]. This spectrum represents a Raman analysis signature, for the characterization of carbon [25,26].

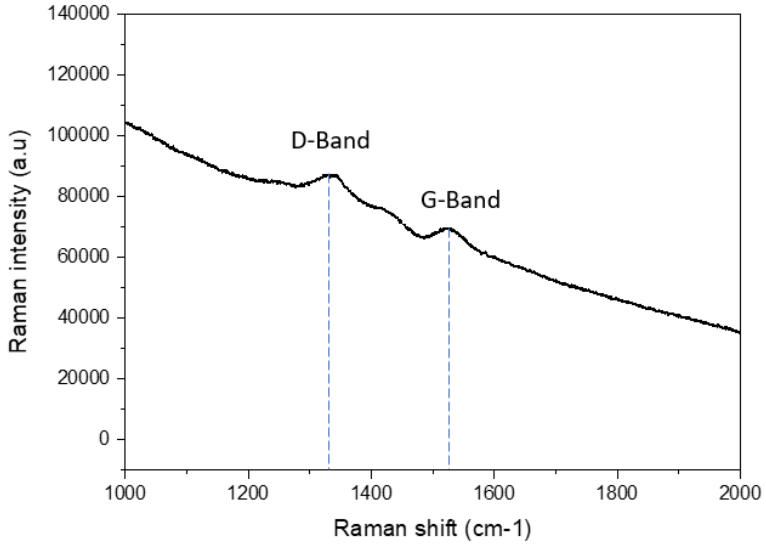

Fig. 4. Raman spectra of pyrolitic carbon in ZTA at 2 hours of holding time of CVI

3.2 Microstructural Analysis

The microstructure of ZTA, with carbon infiltration subjected to different holding times, is shown in Fig. 5. As portrayed in Fig. 5(a), the surface structure porosity of the reference sample without carbon infiltration, is greater than that of the other samples. It was observed that porosity decreased as the holding time was increased.

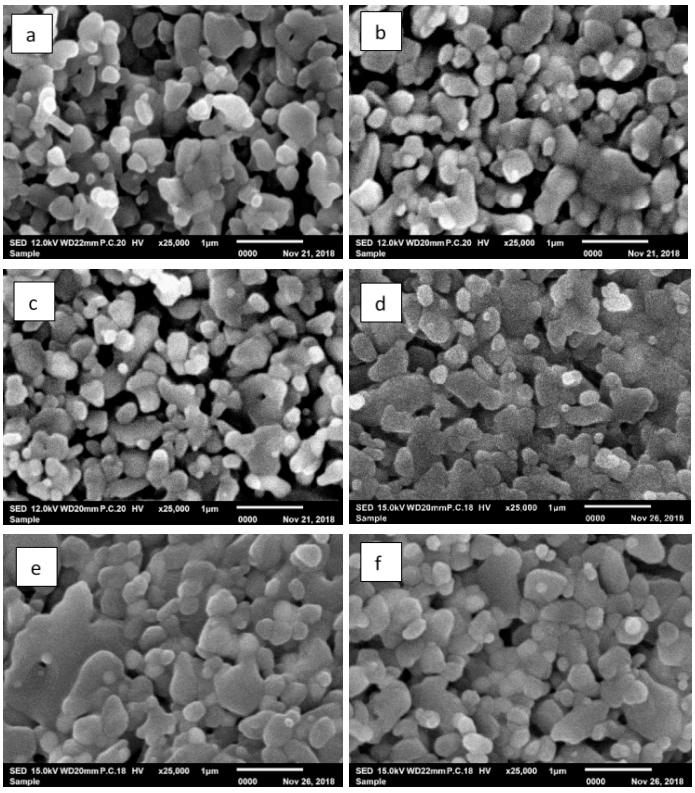

Fig. 5. Microstructure of ZTA-pyrolitic carbon ceramics composite subjected to different holding time of CVI at 25000 magnification (a) 0h, (b) 1h, (c) 1.5h, (d) 2h, (e) 2.5h, (f) 3h

As shown in Fig. 5(d) and 5(e), porosity was significantly reduced, as the pores were filled with carbon during the carbonization process involving tar, as described by Purwanto *et al*. [27]. As reported by Rozhan *et al*. [22], at three hours of holding time, with the decrease in both the tar vapour produced through biomass pyrolysis, and the amount of pyrolytic carbon decomposed into the pore surface, it is safe to assume that carbon infiltration has arrived at the maximum level. This is supported by the BET surface area result, portraying a slight increase in the surface area value, which is an indication of porosity.

3.3 Density

Figure 6 depicts the density of carbon infiltration into the ZTA ceramic composite, at various holding times, during the CVI process. It is apparent that the density of ceramic composite increases in tandem with the increases in holding time. This circumstance could be due to the build-up of carbon in the pores of the composite, as demonstrated by Couto *et al*. [16]. As displayed in Fig. 4(f), the lowest porosity of the microstructure registered at 2.5 hours, contributes towards a high level of density. This high-density level stems from the presence of carbon in the pores, which leads to an enhancement in the mechanical properties of ZTA, as reported by Lee *et al*. [17]. It was observed that the increase in density ended at a holding time of 2.5 hours, before starting to decline from a holding time of 3 hours.

Although the variations in density are insignificant, they serve to verify that carbon infiltration does improve the mechanical properties of ZTA. The insignificant changes in density value, could be due to the minimal infiltration of carbon, into the composite.

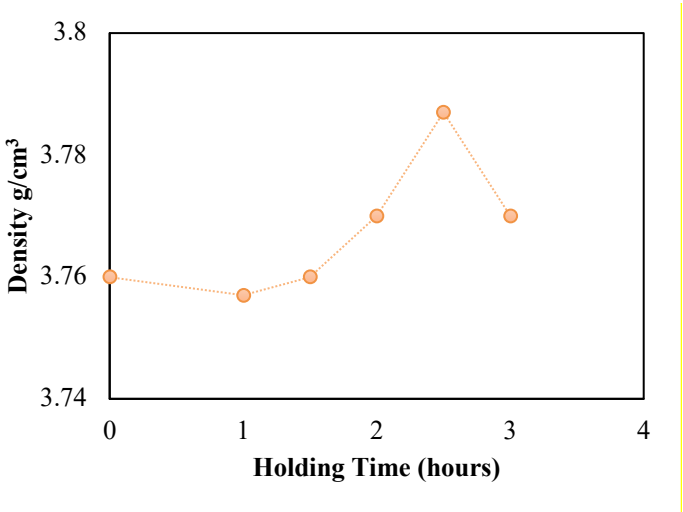

Fig. 6. Density of pyrolytic carbon infiltrated ZTA at different holding time

3.4 Brunauer-Emmett-Teller (BET) and Barret-Joyner-Halenda (BJH) analysis

The BET result for the surface area of samples, for various holding times in the CVI process, is shown in Fig. 7. As can be observed, as the carbon infiltration holding time increased, the surface area of the sample decreased. This finding agrees with that reported in a study conducted by Paek *et al*. [28], in which the surface area and pore volume decreased, with an increase in the carbon load. The decrease, in surface area and pore volume, can be attributed to the infiltration of carbon into the pores. Thus, it can be surmised that a low-porosity solid, has a lesser surface area, than a high-porosity solid.

The surface area drops significantly from 2.1781 m^2/g to 1.8928 m^2/g, as the holding time increased from 1.5 to 2 hours. As shown in Fig. 1, this can be explained by the alteration in XRD intensity, attributed to 2 hours of holding time infiltration of carbon into the ZTA, which led to a reduction in porosity. The lowest surface area value was recorded as 1.6035 m^2/g at 2.5 hours. Any further increment in the holding time, leads to a slight increase in the surface area.

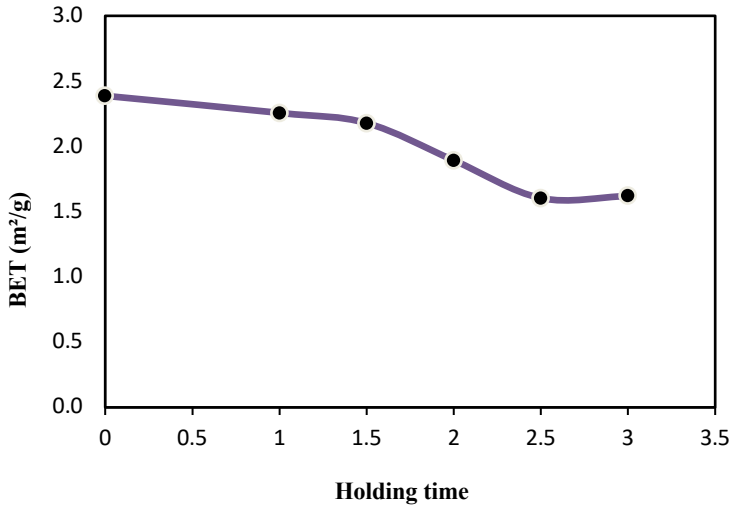

Fig. 7. BET surface area of ZTA-pyrolitic carbon at different holding time

The relationship between pore average width and pore volume, as well as the effect of carbon infiltration on the composite's pore volume is determined through the Barret-Joyner-Halenda (BJH) analysis. As shown in Fig. 8, a decrease in the pore volume triggers an increase in the pore width. According to Hartini *et al*. [29], the decrease in surface area and pore volume of the composite is due to the nature of the composite which tends to approach activated carbon, thus indicating the occurrence of a porosity development activity during the calcination process. Porous reduction is attributable to the growth in microspores, with the increase in activated carbon in the composite.

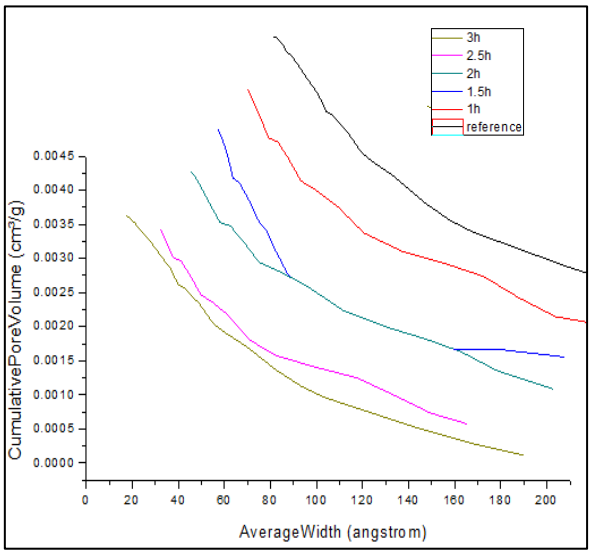

Fig. 8. Pore volume vs average volume

The curve demonstrates that in comparison to a shorter holding time, a longer holding time generates a smaller pore volume. It is predictable that as the holding time increases, more time is available for the pyrolytic carbon to be degraded and distributed within the pore. This is in accordance with the Knudsen diffusion theory, as expressed by Rozhan *et al*. [30].

4. Conclusion

The phase analysis, microstructure, density, and BET of the ZTA with carbon infiltration at different holding times, were investigated. The XRD result revealed a lesser peak intensity at 2 hours of holding time, attributed to the suppression of the ceramic composite's surface, by carbon. The presence of pyrolytic carbon, in the composite, is verified by the appearance of the D peak and the G peak, during the Raman analysis. The optimal holding time of 2.5 hours, is associated to the realization of the least surface area (1.6035 m^2/g), as identified by way of BET analysis. This verifies that the low porosity of the sample is attributable to the high volume of carbon infiltration, into the pores, through the CVI process. The greatest level of density (3.787 g/cm^3) is also achieved at 2.5 hours of holding time. The increase in density, of the ZTA ceramic composite, is due to the build-up of carbon within its pores. However, at a holding time of 3 hours, the density of the sample started to decrease. This indicates that biomass pyrolysis has arrived at its limit. It is essential that the composite be porous, as carbon infiltration is only possible at the surface of its pores. In terms of future work, more emphasis needs to be placed on increasing the porosity of the composite, so that the carbon infiltration process can be enhanced.

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