Microstructure and properties of aluminium-aluminium oxide graded composite materials

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Abstract. In this research works, four-layered aluminium-aluminium oxide (Al- Al₂O₃) graded composite materials were fabricated using powder metallurgy (PM) method. In processing, metal-ceramic graded composite materials of 0%, 10%, 20% and 30% weight percentage of ceramic concentration were prepared under 30 ton compaction load using a cylindrical die-punch set made of steel. After that, two-step pressureless sintering was carried out at sintering temperature and time 600ºC and 3 hours respectively. It was observed that the sintered cylindrical specimens of 30 mm diameter were prepared successfully. The graded composite specimens were analysed and the properties such as density, microstructure and hardness were measured. It was found that after sintering process, the diameter of the graded cylindrical structure was decreased. Using both Archimedes method and rule of mixture (ROM), he density of structure was measured. The obtained results revealed that the microvickers hardness was increased as the ceramic component increases in the graded layer. Moreover, it was observed that the interface of the graded structure is clearly distinguished within the multilayer stack and the ceramic particles are almost uniformly distributed in the Al matrix.

1. Introduction
At present, the demand for multifunctional composite materials is growing increasingly for advanced engineering applications. A new class of graded composite material is suitable for multiple functions and this metal-ceramic graded composite structure can be attained in a single component. Currently, variety of fabrication methods available for processing of these metal-ceramic graded composite materials, namely, centrifugal casting, thermal spraying and powder metallurgy [1-3]. Among these methods, powder metallurgy (PM) is widely used in order to meet the desired requirements and to produce the functional gradients which can also be tailored while providing the best use of each component [4-6]. The most challenging step within powder metallurgy technique is the sintering process. Due to the residual stress caused by the mismatches in thermal expansion and sintering between successive layers, cracks and camber are noticeable during sintering of these multi-layered graded composite materials [7].

In the recent past, several research efforts were carried out to investigate the behaviour of functionally graded materials (FGMs) of different combinations [8-14]. Mechanical responses and the structures of these composites significantly influenced by a number of factors such as powder characteristics, compaction load, metal-ceramic system, sintering temperature, number of layers and the consolidation process. Influence of reinforcements on the mechanical properties of functionally graded aluminium composites was investigated [15]. The obtained results revealed that the outer regions of the composites exhibit higher hardness than the middle and inner regions. The outer region
of all composites showed higher tensile strength than the inner one. Very recently, the effects of various atmospheres on the performances of functionally graded hydroxyapatite/titanium (HA/Ti) were investigated [16]. The obtained results revealed that hydroxyapatite and titanium elements reacted differently depending on sintering atmosphere.

The present research work considers the preparation of four-layered aluminium-aluminium oxide (Al-Al$_2$O$_3$) graded composite specimens using powder metallurgy method. The properties such as densification and shrinkage, microstructure and hardness were analysed.

2. Materials and experimental procedures
In this research, four-layered Al-Al$_2$O$_3$ graded composite samples were fabricated by powder metallurgy route. The Al and Al$_2$O$_3$ powders were mixed with different composition (100:0, 90:10, 80:20, 70:30) according to the design shown in figure 1.

| Composition | Weight Percentage |
|-------------|-------------------|
| 100% Al     |                   |
| 90% Al + 10% Al$_2$O$_3$ |
| 80% Al + 20% Al$_2$O$_3$ |
| 70% Al + 30% Al$_2$O$_3$ |

Figure 1. Composition contribution model of Al-Al$_2$O$_3$ FGM.

Three major steps were involved to process the specimens which are blending, compacting and sintering. At first, the weights of aluminium and aluminium oxide powders were measured based on their molecular weight. After that, these two materials were mixed and blended allowing sufficient time until the good homogenous mixture was achieved. During the stacking process, the powders were filled in order from 70%Al+30%Al$_2$O$_3$ and end with 100%Al (figure 1) into a cylindrical steel die with a diameter of 30 mm. The four-layered powder compositions were cold compacted at 30 ton (294.3 kN) using a hydraulic press (TOYO: Model TL30). After the compaction process, the green compacts are fragile and have low cohesive strengths. Finally, all the composite specimens were consolidated using a sintering furnace (Nabertherm: Made in Germany) by pressureless sintering using a two-step heating cycle/map as shown in figure 2.

Figure 2. Two-step heating cycle for Al-Al$_2$O$_3$ FGM synthesis at sintering temperature 600ºC for 3 hours.

After sintering process, the FGM samples were prepared for characterization and hardness testing. Both green and sintered density of FGM samples were measured by using the rule of mixture (ROM) and Archimedes’ principle respectively. For microstructural studies, the samples were observed by using metallurgical microscope (OLYMPUS BX51M, Made in Japan). The hardness of the FGM
samples was measured using micro-vickers hardness tester (Wilson Hardness: Model 402 MVD, Made in USA) under a load of 300 gf (2.94 N) for a dwell time of 15 seconds along a longitudinal axis with average of 10 measurements.

3. Results and discussion

3.1. Densification and shrinkage

Figure 3 shows the fabricated FGM specimen before and after sintering. The linear shrinkage of Al-Al$_2$O$_3$ graded composite could be calculated accurately by comparing the specimen diameter before and after sintering. The diameter of FGM sample was decreased from 30 mm to 29 mm due to the sintering shrinkage. Neither any physical damage nor any crack was found on above, below or on the sides of fabricated sample. From the obtained results and according to figure 4, the diameter shrinkage of each layer started from 100%Al to 70%Al+30%Al$_2$O$_3$ can be seen decreasing in trend with the increase in Al$_2$O$_3$ from the original diameter 30 mm (green compact).

![Figure 3. Diameter shrinkage due to pressureless sintering.](image)

![Figure 4. Diameter shrinkage each layer of Al-Al$_2$O$_3$.](image)

The densities of the homogeneous layers for different compositions of Al-Al$_2$O$_3$ are shown in figure 5. From the obtained results it can be seen that there is a reduction trend of density with increase in Al$_2$O$_3$ ceramic percentage. After the sintering process, the maximum density was achieved for pure Al layer. It is clear that the reduction trend of density for each homogeneous layer of Al-Al$_2$O$_3$ composite is gradual throughout the system as the ceramic content increases. It is noticed that the sintered density of pure Al layer is slightly higher than the density of green compact. On the other hand, it is apparent that the sintered density of each homogeneous layer of Al-Al$_2$O$_3$ is somewhat lower than the density of green compact.
3.2. Microstructural analysis

The microstructures of sintered Al-Al$_2$O$_3$ graded composites were characterized in order to determine if the particle distribution was consistent and the particle agglomeration occurred. The sintered composites were cut along the cross-sectional direction and mounted using cold mounting process. The mounted specimen was prepared using grinding and polishing. Then the microstructural analysis was carried out using a high resolution optical microscope and the micrographs are shown in figures 6(a)-(d). The microstructure in figure 6 consists of two phases, namely Al and Al$_2$O$_3$ are observed in Al-Al$_2$O$_3$ graded composite materials. The darker phase is Al$_2$O$_3$ and the lighter one is Al matrix. A uniform dispersion of spherical Al$_2$O$_3$ phase is clearly evident in the micrographs for the 2nd, 3rd and 4th layers of the graded structure. It can be seen that the weight percentage of ceramic is increased from 10% to until 30%. Figure 7 shows the interfacial region in Al-Al$_2$O$_3$ graded structure which represents aa’, bb’ and cc’ as the interface layer. No cracking was observed between the layers which indicates that the FGMs have good strength due to the better metal-ceramic bonding and continuous microstructure at the interfaces of the FGMs, as reported in previous studies [12]. From these observations, it is apparent that the interfaces are almost straight and nearly parallel which confirms that powder stacking processes of the graded composites were well executed.

Figure 5. Densities of homogenous layer of Al-Al$_2$O$_3$.

Figure 6. Optical micrographs of Al-Al$_2$O$_3$ graded layers (a) 100% Al (1st layer), (b) 90% Al + 10% Al$_2$O$_3$ (2nd layer), (c) 80% Al + 20% Al$_2$O$_3$ (3rd layer) and (d) 70% Al + 30% Al$_2$O$_3$ (4th layer).
3.3 Microhardness measurement

The variation in hardness across the thickness of graded structure is presented in figure 8. The obtained results revealed that microhardness was increased at each layer with the increase in ceramic content as reported by the researchers in earlier publication [9] and it was noticed that this trend occurred layer by layer almost consistently. From the obtained results, the hardness of layer 1 (100%Al) was measured 29.6 HV. However, at layer 2 (10%Al₂O₃), the hardness was increased to 32.3 HV and further increased to 33.5 HV at layer 3. At layer 4 (30%Al₂O₃), the hardness continued to increase to 35.2 HV as the ceramic content was increased. This increment in hardness was found to be reasonably consistent for dispersion of ceramics in the metal matrix.

4. Conclusions

The reduction trend of density for each homogeneous layer of Al-Al₂O₃ composite is gradual throughout the system as the ceramic content increases. The sintered density of pure Al layer is slightly higher than the density of green compact whereas the sintered density of each homogeneous layer of Al-Al₂O₃ is somewhat lower than the density of green compact. The Al-Al₂O₃ graded composite structure exhibits microstructure with uniform distribution of Al₂O₃ phase in Al matrix. Moreover, the interfacial layers are almost straight and parallel which proves that powder stacking processes and techniques are well executed. The hardness increases with the increase in Al₂O₃ content and this trend occurs layer by layer almost consistently.
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