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RESEARCH ARTICLE

**An ethical committee approval and/or legal/special permission has not been required within the scope of this study.*

PRODUCTION OF TRANSLUCENT ALUMINA CERAMICS BY TAPE CASTING*

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ABSTRACT

Alumina powders are utilized in many industries like electronics, metallurgy optoelectronics and fine ceramic composites. In this study, non-aqueous-based tape casting of alumina was carried out in a non-continuous single blade tape casting machine. A slurry of alumina powder in a solvent was cast on a stationary surface with additives like dispersants, binders and plasticizers. After the green tapes were dried, they were sintered to obtain the final desired shape. Hydraulic pressing method was used to increase the density of the alumina tapes. Layered alumina ceramics were produced at 5 different temperatures by pressureless sintering method (1450°C, 1500°C, 1550°C, 1600°C and 1650°C) for 5 h to observe the effect of sintering temperature. Results revealed the sintering conditions for obtaining translucent alumina, with the utilized slurry composition. Translucent alumina ceramics have gained importance as parts of semiconductor devices, substrates for electric parts, as heat/corrosion materials in reaction tubes and crucibles and are used in medical equipments.

Keywords: *Tape Casting, Alumina, Sintering, Layered Ceramics, Microstructure.*

YARI SAYDAM ALÜMİNA SERAMİKLERİN ŞERİT DÖKÜM YÖNTEMİ İLE ÜRETİMİ

ÖZ

Alümina tozları elektronik, metalurji, optoelektronik ve ince seramik kompozitler gibi birçok farklı endüstride kullanılmaktadır. Bu çalışmada, alüminanın su bazlı olmayan şerit dökümü, sürekli olmayan tek bıçaklı bir şerit döküm makinesinde gerçekleştirilmiştir. Dağıtıcılar, bağlayıcılar ve plastikleştiriciler gibi katkı maddeleri içeren bir çözücü içindeki alümina tozundan oluşan bir süspansiyon, sabit bir yüzey üzerine dökülmüştür. Ham şeritler daha sonra kurutulmuş ve son olarak istenen nihai şekli elde etmek için sinterlenmiştir. Alümina ham şeritlerin mikro yapısını optimize etmek ve yoğunluğunu artırmak için hidrolik presleme yöntemi ile preslenmiştir. Katmanlı alümina seramikler, sinterleme sıcaklığının mikroyapı üzerindeki etkisini gözlemlemek için 5 farklı sıcaklıkta (1450°C, 1500°C, 1550°C, 1600°C ve 1650°C) 5 saat süreyle basınçsız sinterleme yöntemiyle üretilmiştir. Sonuçlar, kullanılan süspansiyon bileşimi ile yarı saydam alümina elde etmek için sinterleme koşullarını ortaya çıkarmıştır. Yarı saydam alümina seramikler yarı iletken aygıtlarda, devre altlıklarında, ısı ve korozyona dayanıklı malzeme olarak reaksiyon tüpleri ve potalarda, medikal uygulamalarda kullanım alanı bulmaktadır.

Anahtar Kelimeler: *Şerit Döküm, Alümina, Sinterleme, Tabakalı Seramikler, Mikroyapı.*

1. INTRODUCTION

Alumina is utilized in many applications due to its thermal and chemical stability, high wear and corrosion resistance, high hardness and strength and excellent electrical insulation properties **(**Fujiwara et al., 2007; Vuksic et al., 2021a; Yu et al., 2015; Vuksic et al., 2021b; Manotham et al., 2021). However, being a ceramic material, its inherent brittleness causes low mechanical stability and reliability and limits its use. Thus, many studies are being carried out to improve the toughness of alumina ceramics.

One of the methods to enhance reliability of such ceramics is production of laminated structures. Tape casting has been developed to produce ceramic tapes used in these laminated structures (Mortara, 2005). The process involves casting a specially formulated slurry onto a stationary or moving surface. The slurry involves ceramic powder in solvent having other additives like binders, dispersants, and plasticizers (Hou et al., 2019; Zhao et al., 2020). Resulting green body has better uniformity compared to traditional dry pressing since it is formed by in situ consolidation of polymers (Jurkow et al., 2011; Liu et al., 2012; Ba et al., 2013; Naebe & Shirvanimoghaddam, 2016; Vozdecky et al., 2010; Feng et al., 2020). Besides, large areas of thin sheets with controlled thickness, within the range of 10 µm to 1 mm, can be produced by this method with low cost (Mistler, 1995). Although pores and discontinuities between the layers present a problem, there are many ongoing studies to improve this production technique (Miao & Sun, 2010).

Ceramic tape technology provides means to create complex 3D structures, thus has a great potential for production of many devices (Roosen, 2006). It is also possible to produce functionally graded materials with tape casting, having dense outer layers and porous inner layers. The method is utilized for production of translucent polycrystalline alumina as well, which is the preferred material for arc tubes in high-pressure sodium (HPS) discharge lamps (Zhou et al., 2016, Pabst & Hříbalová, 2021; Wei et al., 2001; Willems, 1992).

The present study is carried out with the aim to determine the process parameters in tape casting for production of translucent alumina. A well-designed slurry composition is used for this purpose and the effect of sintering temperature on alumina ceramic thin layers is evaluated. Although translucent and transparent ceramics have six decades of history, extensive research is still being carried out on the development of these materials.

2. EXPERIMENTAL STUDIES

Alumina powder used as a raw material in this study, A-16 SG from Alcoa with 99,8 % purity, was characterized first in terms of particle size distribution and phases present. X-ray diffraction studies using Shimadzu XRD 6000 diffractometer revealed that alumina powder consisted of single phase of alumina (corundum). Average particle size of the powder was 300 nm as determined by laser particle size analyzer, Malvern Zetasizer NanoZS 3600. Microstructures of both alumina powder and sintered body were observed using JEOL 633F scanning electron microscope (SEM). Figure 1 shows the SEM image of α-alumina powder used in the study. In the suspension preparation process, MEK (Merck KGaA), EtOH (Merck KGaA) were used as solvent media, PVB (Chang Chun Petrochimical Co. Ltd.) was used as solvent-based binder, STPP (Eczacıbaşı) was used as dispersant, DBF (Plastifay Chemical Industry Co.) and PEG400 (Merck KGaA) were used as plasticizers.

The slurry composition was formulated to provide a good dispersion of ceramic and to obtain a stable suspension so that a homogeneous and compact green structure could be produced. The organic additives were selected to enhance the tape characteristics, without interacting. The binder system was chosen such that the mechanical characteristics of the tape would be improved without compromising the green density or inducing flocculation.

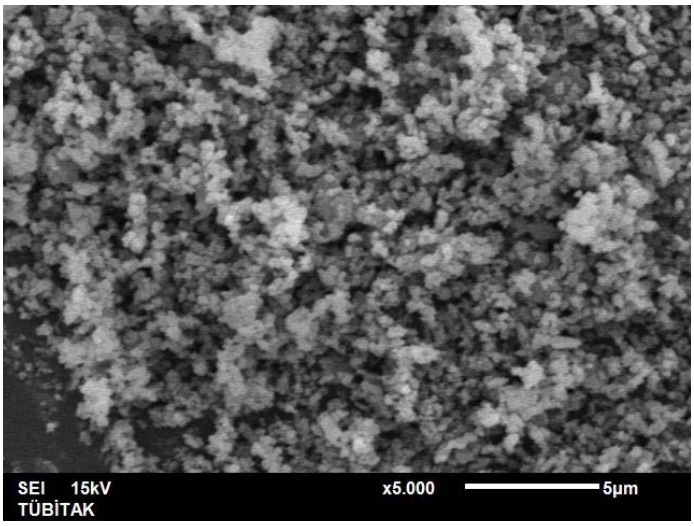


Figure 1. SEM image of the alumina powder.

The slurry was used in a non-continuous single blade tape casting machine to produce the green alumina tapes by non-aqueous alcohol based tape casting method. After drying the green tapes were sintered at temperatures 1450°C, 1500°C, 1550°C, 1600°C and 1650°C for 5 hours each. Preparation of the tapes and the layered structure is shown schematically in Figure 2, the slurry was spread by hand motion.

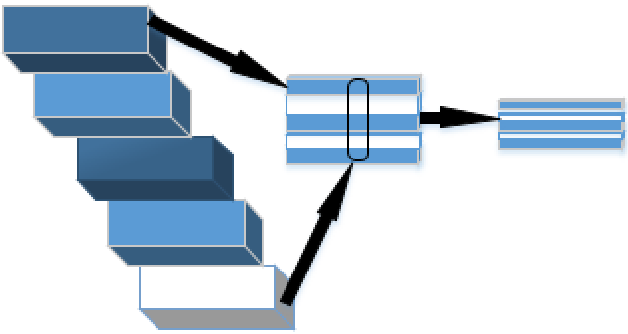


Figure 2. Preparation of the tapes and the layered structure.

Dispersion studies were carried out with ~ 50 wt % of solid content. Suspensions were prepared by dispersion of the alumina powder in MEK/EtOH based solvent system. First stage of the preparation was deagglomeration of alumina powder and its dispersion in the solvent. The solvent was an azeotropic mixture of 64 vol % MEK and 36 vol % EtOH. Second stage involved mixing of the solvent with the binder, dispersant and plasticizers. Ball milling was carried out by mixing 100 g slurry with 125 g zirconia balls for 24 h, for effective deagglomeration and dispersion. The viscosity was measured as 63 centipoise (cP) using Brookfield LVDV-II+P viscometer.

Tape casting was carried out using doctor blade technique on a clean glass bed using a laboratory tape caster (MSE Teknoloji Ltd., TR). The blade gap was kept at 250 μm. The cast tapes were dried at ambient conditions for 12 hours. After drying, tapes were released and inspected for potential defects.

Laminates were prepared by stacking 5 sheets manually, that were 2x1 cm in dimensions. They were then compressed by hydraulic press (PMK brand model) at 200 MPa. Sintering was carried out in a Protherm PLF160/63 furnace at air atmosphere at different temperatures (1450 °C, 1500 °C, 1550 °C, 1600 °C and 1600 °C) for 5 h each. A heating rate of 5 °C/min was used and furnace cooling was adopted. Thicknesses of the thin ceramics were measured and firing shrinkage values were calculated after sintering. Illustration of the whole tape casting process is given in the Figure 3.

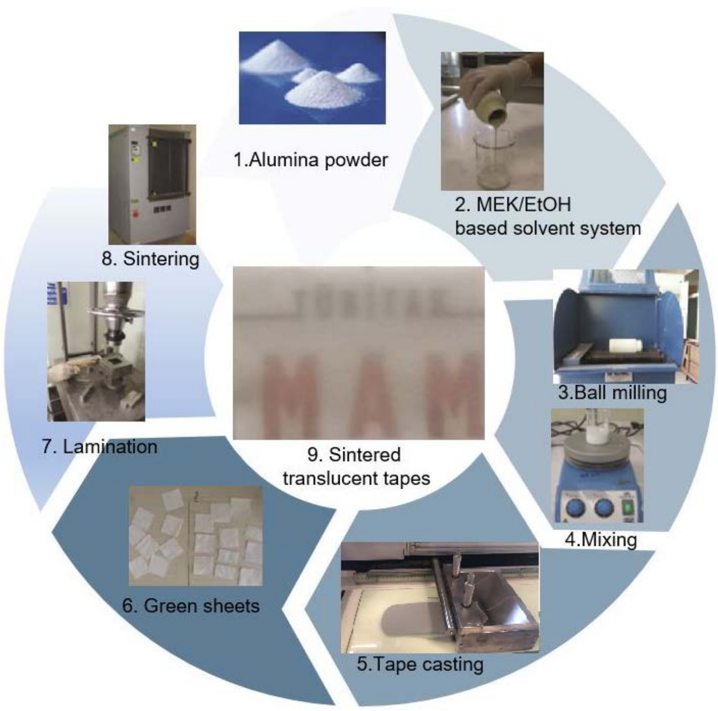
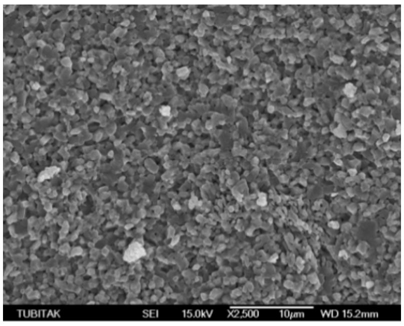


Figure 3. Illustration of the whole tape casting process.

3. RESULTS AND DISCUSSION

The microstructures of the sintered tape samples, examined using SEM at 2500X, are shown in Figure 4. Effect of sintering temperature on the grain shape and size can be clearly seen in these images. As the sintering temperature increases the grain size increases and porosity decreases indicating a denser structure. The SEM results are supported by the calculated theoretical density values of the samples which increased from 83,3 % (1450 °C) to 90 % (1650 °C). Theoretical densities were determined according to Archimedes principle and used in calculation of firing shrinkage values. For the samples sintered between 1450-1550 °C shrinkage was 16-17 %, whereas it was between 17-18 % for those sintered between 1600-1650 °C.



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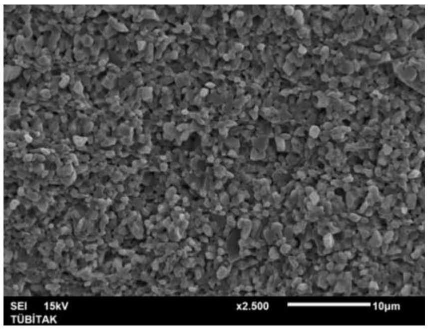

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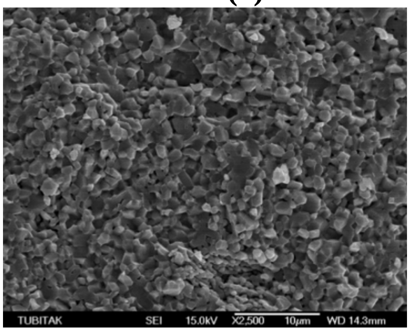

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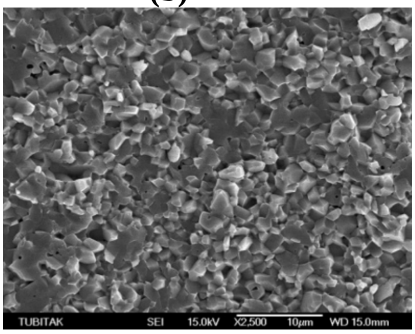

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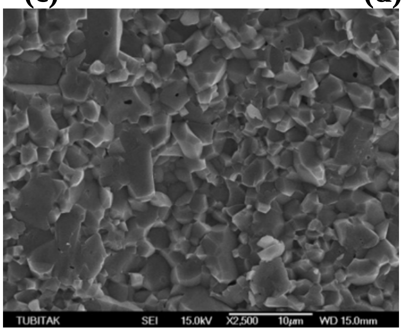
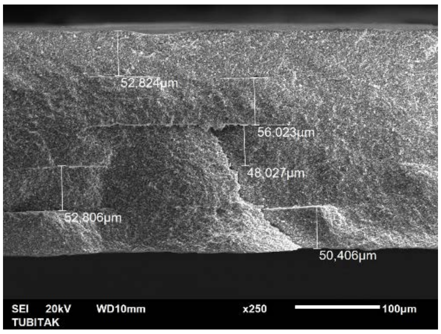


(e)

Figure 4. The microstructures of the sintered tape samples a) 1450 °C, b) 1500 °C, c) 1550 °C, d) 1600 °C, e) 1650 °C.



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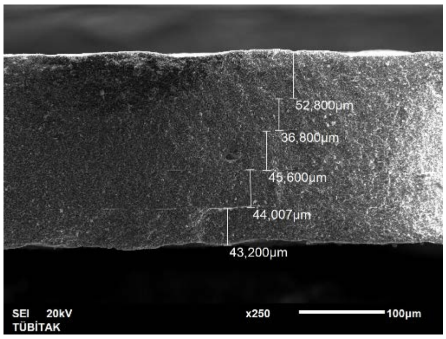

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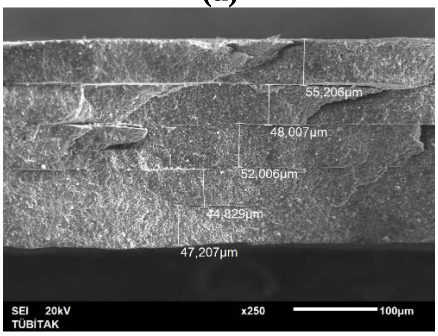

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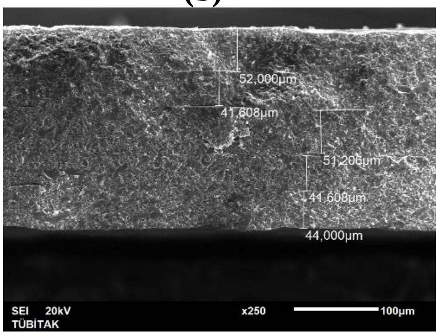

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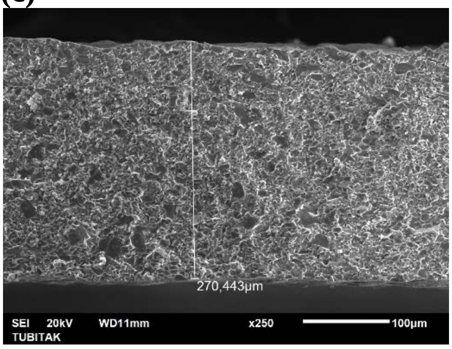

(e)

Figure 5. The cross sections of the alumina tapes sintered at different temperatures a) 1450 °C, b) 1500 °C, c) 1550 °C, d) 1600 °C, e) 1650 °C**.**

The cross sections of the alumina tapes sintered at different temperatures were also analyzed by SEM and layer thicknesses were determined (Figure 5).

As seen from the figures, sintering at temperatures between 1450-1600 °C revealed distinctly all five layers, with thicknesses varying between 36-52 µm (Figure 5a-d). However, at sintering temperature 1650 °C layers cannot be distinguished from one another resulting in the densest structure. A translucent alumina was actually produced by sintering at 1650 °C as shown in Figure 6.

Figure 6. Five layer tapes sintered at 1650 °C.

It is known that the sintering temperature for alumina densification usually increases with the increase in its purity and if high purity alumina powders are prepared by the traditional sintering method, the sintering temperature should be over 1700 °C to get a dense alumina sintered body (Verma & Kumar, 2016; Figiel et al., 2011). In this study, it is shown that with a well-designed slurry composition and high purity singlephase powder, it is possible to obtain a dense and translucent alumina at a lower sintering temperature than 1700 °C. Achieving a dense structure at lower temperatures helps also to avoid abnormal grain growth of alumina, which leads to loss in flexural strength, and wear resistance.

4. CONCLUSION

In this study alumina tapes were prepared by tape casting method and layered structures were obtained by pressureless sintering method using 5 different temperatures (1450°C, 1500°C, 1550°C, 1600°C and 1650°C) and 5 hours holding time, to observe the effect of sintering temperature on the microstructure. SEM studies revealed that as the sintering temperature increased the grain size increased and porosity decreased, indicating a denser structure. Theoretical density values increased from 83,3 to 90 % by increasing the sintering temperature from 1450 to 1650 °C, supporting the SEM studies. Theoretical densities were used in calculation of firing shrinkage values. For the samples sintered between 1450-1550 °C shrinkage was 16-17 % whereas it was between 17-18 % for those sintered between 1600-1650 °C. The cross sections of the layered structures were also studied by SEM and the sample sintered at the highest temperature revealed the most compact structure having translucent characteristics. This study has shown that it is possible to produce translucent alumina ceramics by tape casting without vacuum sintering process.

CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

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