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Tape Casting of Magnesium Oxide

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Tape Casting of Magnesium Oxide

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ABSTRACT

A tape casting procedure for fabricating ceramic magnesium oxide tapes has been developed as a method to produce flat sheets of sintered MgO that are thin and porous. Thickness of single layer tapes is in the range of 200-400 μm with corresponding surface roughness values in the range of 10-20 μm as measured by laser profilometry. Development of the tape casting technique required optimization of pretreatment for the starting magnesium oxide (MgO) powder as well as a detailed study of the casting slurry preparation and subsequent heat treatments for sintering and final tape flattening. Milling time of the ceramic powder, plasticizer, and binder mixture was identified as a primary factor affecting surface morphology of the tapes. In general, longer milling times resulted in green tapes with a noticeably smoother surface. This work demonstrates that meticulous control of the entire tape casting operation is necessary to obtain high-quality MgO tapes.

ACKNOWLEDGMENTS

The authors thank John S. Stuecker of the Ceramic and Inorganic Materials Department (1815) for sharing his expertise in ceramic slurry preparation and for numerous conversations regarding the tape casting process.

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NOMENCLATURE

Al ₂ O ₃	Alumina
BET	Brunauer, Emmett, and Teller
BBO	Binder burnout
BBP	Butyl benzyl phthalate
HDPE	High density polyethylene
MEK	Methyl ethyl ketone
MgO	Magnesium oxide
SEM	Scanning electron microscopy
SSA	Specific surface area
TGA	Thermogravimetric analysis
ZrO ₂	Zirconia

1. INTRODUCTION

A need for thin, flat sheets of magnesium oxide with high levels of porosity after sintering motivated a process development study using tape casting, a class of slurry forming techniques. Tape casting or doctor blading is a fluid forming technique that is widely used for large-scale fabrication of thin ceramic substrates that are virtually impossible to press and extrude. Green, or unfired, ceramic sheets are flexible and can be cut into a variety of shapes and sizes depending on the specific application. Typical thicknesses for the green tapes range from about 20 μm to more than 1 mm. The tape casting technique consists of depositing a thin layer of a slurry or slip onto a moving surface using a "doctor" or scraping blade to remove excess slurry from the surface being coated (1). Prior to tape casting, the height of the doctor blade is set to a desired tape thickness. A schematic diagram of the tape casting experimental set up is shown in Figure 1 and the actual laboratory scale tape casting set-up used in this study is shown in Figure 2.

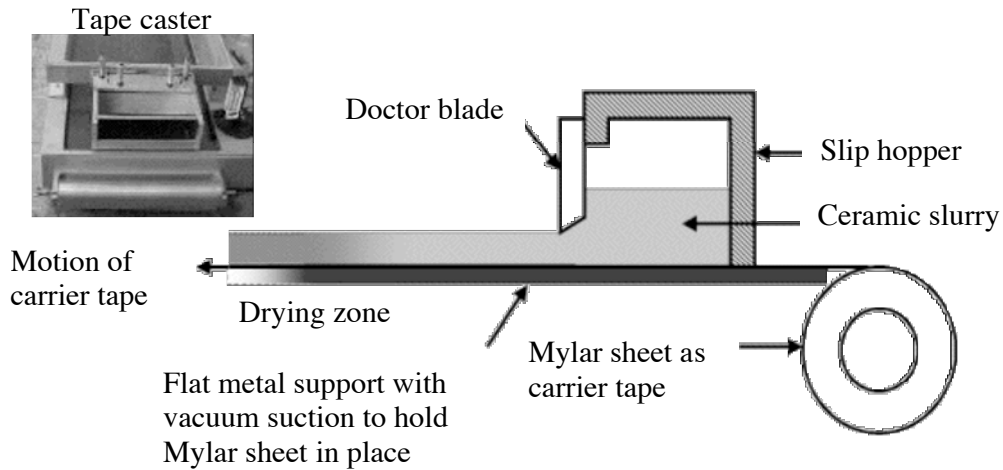


Figure 1. Schematic diagram of the tape casting process. The ceramic slurry is drawn out beneath the doctor blade by the motion of the carrier. The green tape thickness is controlled by the preset height of the doctor blade.

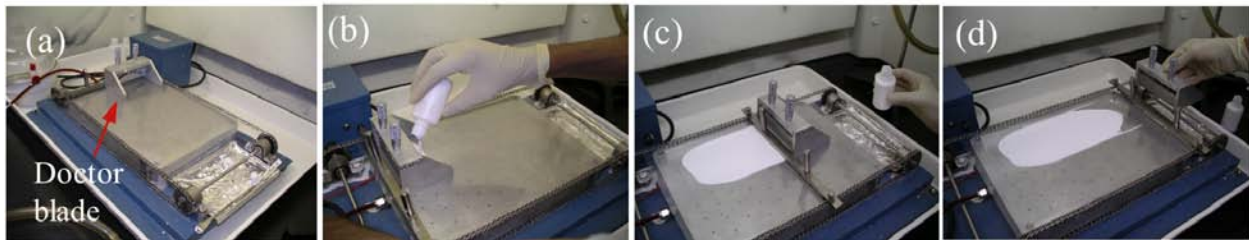


Figure 2. The tape casting process. (a) Tape caster used in this study. (b) The slurry or slip is initially poured onto the casting surface. (c) The moving doctor blade removes excess slurry, leaving behind a tape with a preset thickness. (d) The as-cast tape is left to dry overnight.

The quality of tape cast ceramics depends on two processing parameters: (1) ceramic slip preparation and (2) firing schedules. Slips are multicomponent systems consisting of a powder (ceramic, metallic, or composite), a solvent, a dispersant, a binder, and a plasticizer. Each component plays a specialized role in the tape casting process. The solvent is the carrier medium used to dissolve and homogeneously distribute the other slip components. The dispersant or deflocculant disperses the particles in the system to keep them apart and homogeneously suspended in the slip. The polymeric binder supplies the network that holds the entire chemical system together. A green tape can be accurately described as a polymer matrix impregnated with large quantities of a ceramic material. The plasticizer is added to the tape casting slip to add flexibility to the green tape so that it may be handled without cracking. Of these components, the powder, a ceramic powder in this case, is the most important ingredient in the batch formulation. After sintering of the green ceramic tapes, the powder is the only portion of the slip that is left behind to define the properties of the manufactured part. The solvent, dispersant, binder, and plasticizer facilitate the fabrication of the ceramic tape but are lost during firing for binder burnout (BBO) and the final sintering.

Powders are selected for processing by tape casting based on desired properties of the final ceramic product, for example grain size or phase purity. The starting powders must be well characterized to have a better control of the numerous variables involved in tape casting. The important parameters to monitor in all powder batches are average particle size and distribution, surface area, and trace-impurity level.

As-received powders are typically agglomerated or consist of groups of particles that stick together. Accurate particle size distributions and particle shapes are obtained by milling the powder to break up agglomerates. Calcination, or heating of the powder to high temperatures, is the typical processing step prior to characterizing powders for size distribution and particle shape. (Figure 3 presents a process flowchart for the steps involved in tape casting.) Calcination of the as-received powders cleans the powder by driving off water present as absorbed moisture, driving off carbon dioxide, and/or oxidizing/reducing the powder to the desired state. Typically, powders are heated to several different temperatures to determine the optimum conditions for calcination. Powder calcination will also affect the surface area. In general, powders with surface areas in the 5 to 15 m²/g range (as determined by the BET method) are much easier to work with. In preparing a powder for tape casting, a processing compromise must be reached between milling, calcination, and a favorable surface area.

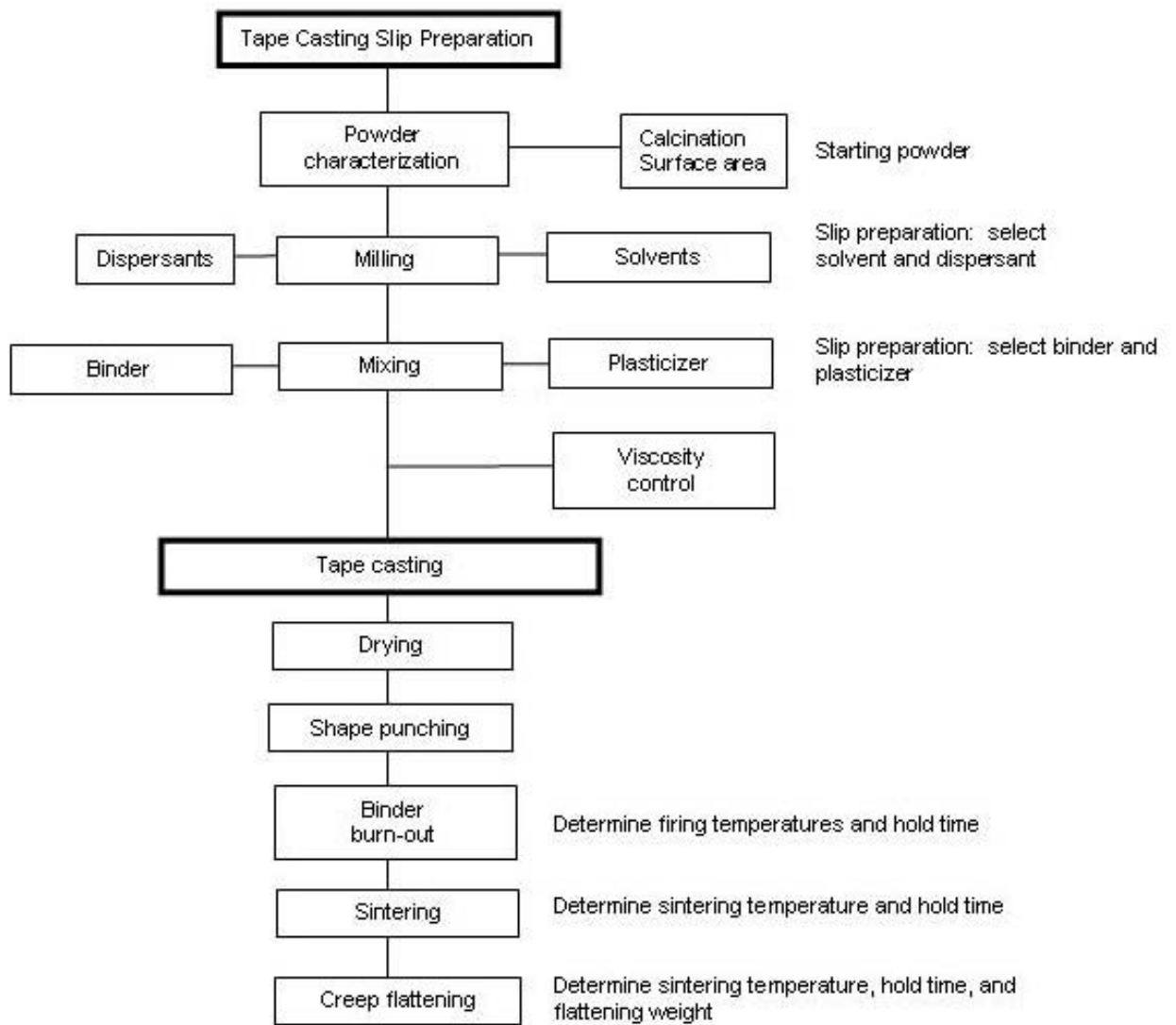


Figure 3. Process flowchart for tape casting.

2. TECHNICAL APPROACH

2.1 Starting Material and Calcination Treatment of MgO Powder

Chemical solution precipitation methods are commonly used to produce high purity ceramic powders (2). The resulting material is calcined to convert the starting mixture into a pure oxide compound. The development of a tape casting technique for fabricating MgO tapes required a parallel calcination study to investigate how to prepare sinterable ceramic powders with particle size and surface area suitable for tape casting. In order to tape cast MgO powder into thin tapes (~300 μm), the starting particle size had to be smaller than the final sintered thickness of the tape and the specific surface area (SSA) had to be $< 20 \text{ m}^2\text{g}^{-1}$ in order to handle and process the nonaqueous slip reproducibly.

MgO powder from Inframat Advanced Materials LLC (Farmington, CT, USA) was selected for this study based on manufacturers' data on purity and particle size. The MgO powder was reportedly made using Inframat's proprietary wet chemical precipitation method, followed by a calcination treatment. The as-received powder was stated to have the following powder characteristics:

Particle size	0.5 - 2.0 μm
SSA	$< 20 \text{ m}^2\text{g}^{-1}$
Density	3.60 g cm^{-3}

The Inframat MgO powder is 99.9% pure MgO with the minor impurities listed in Table 1. The high purity designation pertains only to metallic impurities. Based on TGA measurements, the pure oxide seems to have significant residual carbon and hydrogen on the surface of the powder from the precipitation method.

Table 1. Inframat MgO (99.0% pure) metal impurities.

Impurities (wt%)	Max Level
Al_2O_3	0.02
B_2O_3	0.0002
CaO	0.05
Fe_2O_3	0.001
MnO_2	0.001
Na_2O	0.001
NiO	0.0005
SiO_2	0.001
ZnO	0.0005

In order to optimize the Inframat MgO powder properties for tape casting, a series of calcination heat treatments was performed at 400, 600, 800, 1000, 1200 and 1400 $^{\circ}\text{C}$ for four hours each. Calcining at higher temperatures causes sintering of the MgO crystallites, resulting in a general decrease in surface area and increase in particle size. These results are shown in Figure 4. Calcining at 1200 $^{\circ}\text{C}$ for four hours was found to result in a SSA and particle size that were adaptable to tape casting, $14 \text{ m}^2\text{g}^{-1}$ and 1.5 μm , respectively. Figure 5 shows scanning electron micrographs for the MgO powders at each calcination temperature.

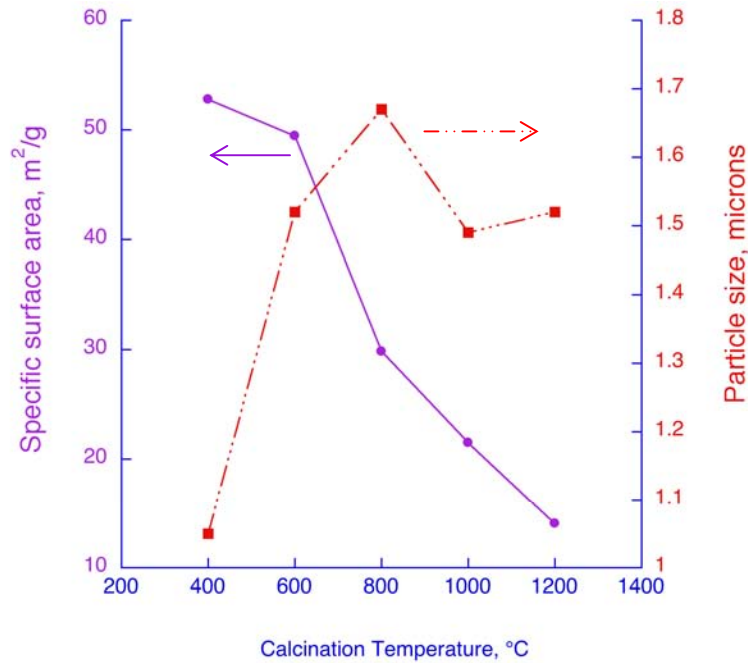


Figure 4. Specific surface areas and average particle sizes for Inframat MgO powder calcined at 400, 600, 800, 1000 and 1200 °C. SSA and particle size measurements were conducted using standard BET N₂ (g) adsorption and dynamic light scattering techniques.

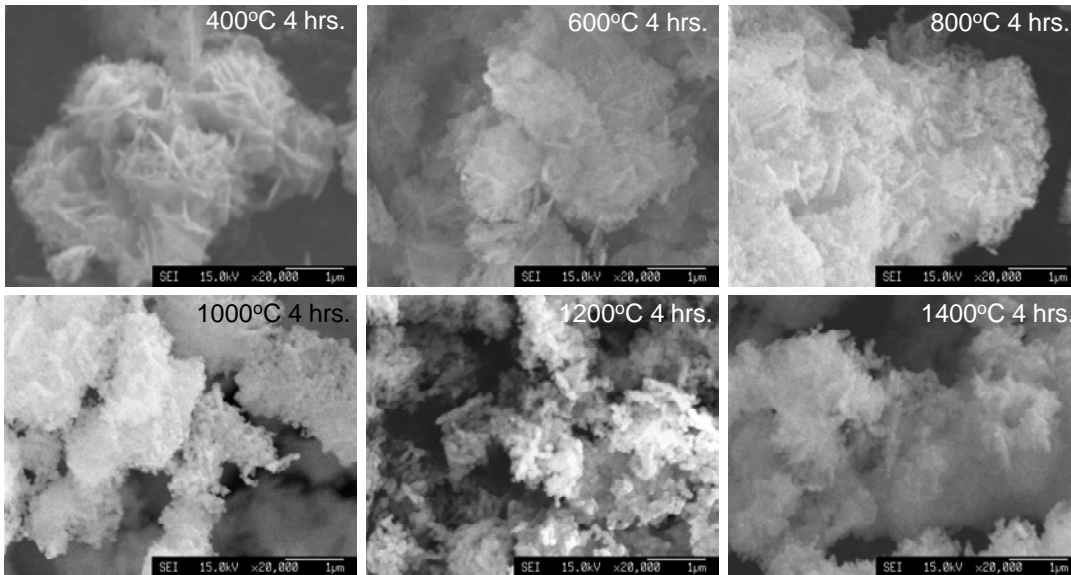


Figure 5. Scanning electron micrographs for Inframat MgO powder calcined at 400, 600, 800, 1000, 1200 and 1400 °C for 4 hours. The fine crystallite structure at 400 °C densifies with increased calcinations temperature. Therefore, the surface area decreases and particle size increases with increasing temperature as a result of the early stages of sintering.

2.2 MgO Powder Dispersion Study

Powder pre-treatment conditions for the tape casting preparation were determined using SEM and BET characterization of the as-received MgO powder. The next step was to achieve uniform and homogeneous dispersion of the ceramic powder in the slip. Tape casting slips must be well dispersed in order to achieve a high packed-bed density in the green tape. A study was designed to observe the dispersion behavior of identical suspensions of powder and solvent prepared with different concentrations of dispersant. Based on previous experience in tape casting, methyl ethyl ketone (MEK)/toluene was chosen as the solvent. A fish oil dispersant was initially used for slurry preparation, but did not effectively disperse the MgO. Homogeneous dispersion was obtained with Solsperse 2400SC dispersant (Avecia Corp.). Five slips were prepared with like amounts of MgO (10 g) and solvent (10 g), but varying amounts of the dispersant. The dispersant was added as 1, 2, 3, 4, and 5 weight percent to each suspension. The suspensions were rolled on a mill for 4 hours. Ten mL of each suspension was poured into 10 mL graduated cylinders, sealed with Parafilm, and allowed to sit undisturbed for 3 days. After 3 days, the packed-bed heights were recorded. Figure 6 shows that maximum settling, and therefore best effective dispersion, was obtained with 3 weight % additions of Solsperse 2400.

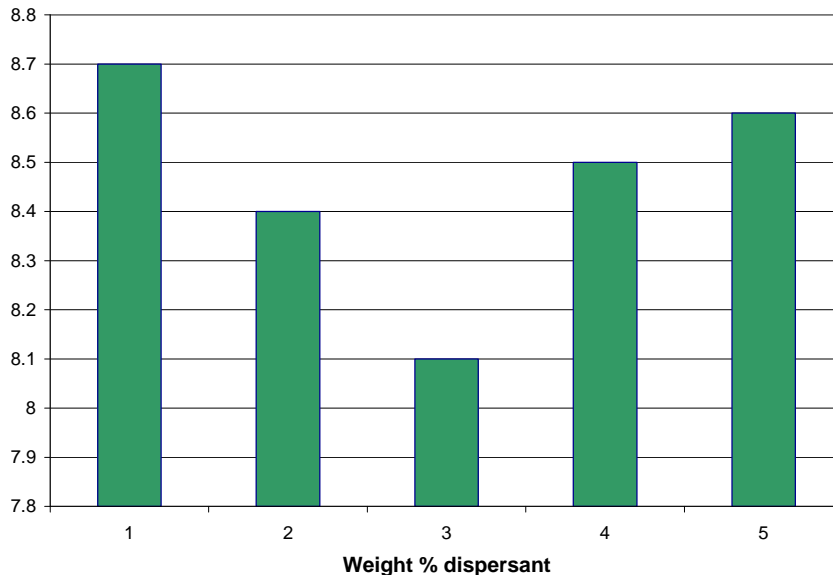


Figure 6. Results of dispersion study on MgO suspensions prepared with MEK/toluene as the solvent and Solsperse 2400 dispersant.

2.3 Tape Casting Procedure

2.3.1 Slurry Preparation

Tape casting slurries were prepared by dissolving a Solsperse 2400SC dispersant (Avecia Corp.) in a solvent mixture of MEK and toluene in a 1:1 ratio by weight. The MgO powder (Inframat) was added and one ZrO₂ milling ceramic ball was placed in the mixture to aid in the mixing and dispersion. In some experimental runs, high density polyethylene particles (HDPE) (average particle size of 18 μm [HD-1800; Inhance Fluoro-Seal Corp., Houston, TX, USA]) were added at this stage to assess their efficacy in increasing the porosity of the sintered sheets.

The mixture was milled for 10 minutes in a Model 8000 M Mixer/mill (SpexMill Industries Inc., Edison, NJ). In the second milling step, butyl benzyl phthalate (BBP) as a plasticizer and Butvar B-79 Polyvinyl Butyral Resin (B79) as the binder were added to the suspension and milled for 10 minutes. The slip was milled in a rolling ball mill for a minimum of 48 additional hours. Ball milling times in excess of 96 hours resulted in green tapes with a noticeably smoother surface. Tables 2 and 3 provide the detailed compositions of the MgO slurry and the MgO slurry with HDPE pore former. Both compositions are based on batches using 10 g of MgO powder. As can be seen in Table 2, the composition of the MgO slurry is given by the ratios:

- 2 wt% dispersant: MgO
- 20 wt% binder: MgO
- 10 wt% plasticizer: MgO
- 15 volume % MgO

The composition for the MgO slurry with the HDPE pore former is given by the ratios:

- 10 volume % HDPE: MgO
- 1.9 wt% dispersant: (MgO+HDPE)
- 19.5 wt% binder: (MgO+HDPE)
- 9.7 wt% plasticizer: (MgO+HDPE)
- 16.9 volume % MgO and HDPE

After milling, the slip was degassed in a bell jar held under vacuum (10^{-2} Torr) for 20 seconds. The slip was poured onto a Mylar sheet set on a bench top tape caster (Gardner Laboratory, Silver Spring, MD) and spread as a thin layer with a casting speed of 1 cm/sec and a doctor blade adjusted to a height of approximately 0.6 mm. The tape was allowed to dry overnight. The green tapes had a tendency to develop cracks during drying under vacuum. Small discs with a diameter of 28.5 mm were cut from the uniform portions of the tapes using a circular brass punch and a hammer.

Table 2. MgO Slurry Composition

	Materials	Density, g cm ⁻³	Volume Percent	Mass Percent
Dispersant	Solsperse 2400	1.13	0.98	0.85
Solvent	MEK	0.8	34.62	21.31
Solvent	Toluene	0.86	32.20	21.31
Plasticizer	BBP	1.1	5.04	4.26
Binder	Butvar B-79	1.083	10.23	8.53
Powder	MgO	3.6	15.39	42.63

Table 3. MgO and Pore Former Slurry Composition

	Materials	Density, g/cc	Volume Percent	Mass Percent
Dispersant	Solsperse 2400	1.13	0.98	0.85
Solvent	MEK	0.8	34.62	21.31
Solvent	Toluene	0.86	32.20	21.31
Plasticizer	BBP	1.1	5.04	4.26
Binder	Butvar B-79	1.083	10.23	8.53
Powder	MgO	3.6	15.39	42.63
Pore Former	HDPE	0.93	1.55	1.11

2.3.2 Binder Burnout

Removing the binder and other organics from the green tape is usually a lengthy process that ensures the tapes are ready for high temperature densification during sintering. The tapes are highly loaded with binder in order to give them flexibility and an appropriate BBO cycle is needed. The temperature cycle for binder and pore former removal in air was determined using thermogravimetric analysis (TGA) on the pure organics. Figure 7(a) shows that the temperature for Butvar B-79 removal begins at 425°C. The temperature where most of the pore former, HDPE, is removed in air starts at 450°C, as seen in Figure 7(b). Since the temperature for removal of the binder and HDPE are at 600 and 500°C, a conservative BBO cycle should go higher than these temperatures to ensure complete removal of binder and pore former. It is important to note that the tapes are partially sintered during the BBO cycle at 1050 °C to give them enough strength for handling. Based on the binder burnout experiments, the optimized BBO cycle for MgO and MgO-HDPE green tapes is shown below.

1. Heating: 3°C/min. up to 425°C hold for 60 min.
2. Heating: 3°C/min. up to 1050°C hold for 120 min
3. Cooling: 5°C/min. down to 100°C then 20°C/min. down to room temperature

The final sintering step requires a flat, smooth, non-adherent surface to support the ceramic tapes. For initial experiments, some tapes were burned out and sintered on an untreated Al₂O₃ lattice, on an Al₂O₃ lattice sprayed with boron nitride to prevent sticking, and on powder beds of Al₂O₃ or MgO. The tapes adhered to the untreated or pretreated Al₂O₃ lattice, resulting in severe cracking. Both of the powder beds prevented adhesion and cracking of the tapes, but resulted in tapes that sintered with the texture of the powder bed and were wavy.

Better surface quality was achieved by using rectangular 1 in. by 1 in. alumina substrates placed in a tile-like arrangement in an alumina (Al₂O₃) crucible to provide a setter for the tapes during BBO and sintering. The surface of the substrates was pretreated with a brushed-on coat of an aqueous zirconia (ZrO₂) suspension (dried at room temperature) to prevent the tapes from sticking. The ZrO₂ coating also provided a flat, uniform surface for tape sintering. The green tape discs were placed on the ZrO₂ coated Al₂O₃ substrates with the Mylar side down (the side in contact with the Mylar during tape casting). The crucible was placed in a bench-top Thermolyne F48000 air furnace (Barnstead International) to burn out the binder, the pore former (if present), and other organics.

2.3.3 Sintering

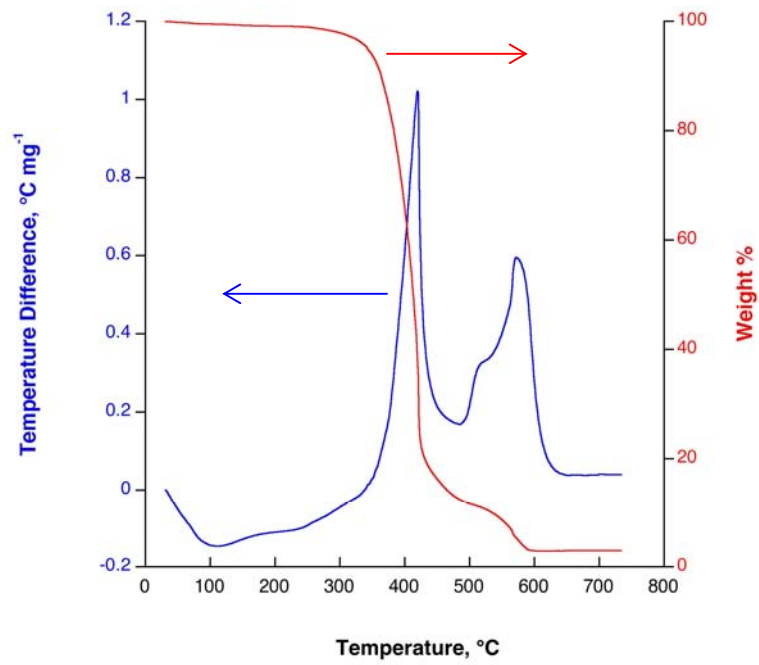
The MgO tapes were sintered in a high temperature box furnace (Sentro Tech Corp, Cleveland, OH) at 1450°C for different hold times. The firing schedule is shown below.

1. Heating: 10°C/min. up to 1450°C hold for 120 min. or 240 min.
2. Cooling: 10°C/min. down to room temperature

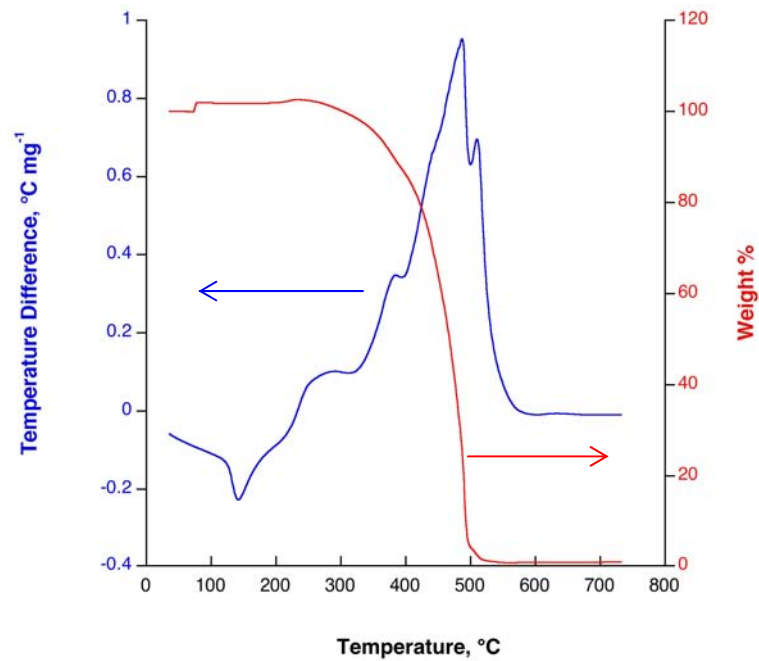
2.3.4 Creep Flattening

Post-sintering firing was required to reduce tape curling. Three Al₂O₃ substrates (1.5 g each) were placed on top of the as-sintered MgO tapes to provide weight for tape flattening, a procedure usually referred to as creep flattening. The surface of the Al₂O₃ substrate in contact with the tapes was coated with ZrO₂ to prevent sticking. The firing schedule is shown below.

1. Heating: 10°C/min. up to 1500°C hold 30 min.
2. Cooling: 10°C/min. down to room temperature



(a)



(b)

Figure 7. TGA-DTA analysis shows the (a) B-79 binder and (b) HDPE pore former begin to burn off in air at 400 °C and at 450 °C, respectively. A hold at 425 °C allows for the diffusion of most of the gaseous products out of the tapes.

3. CHARACTERIZATION OF MgO CERAMIC TAPES

3.1 Apparent Porosity and Density Measurements

Densities of the green and sintered MgO tapes were determined using ASTM Standard C830-00, "Standard test methods for apparent porosity, liquid absorption, apparent specific gravity, and bulk density of refractory shapes by vacuum pressure". According to this standard, apparent porosity and specimen density are calculated by measuring the dry weight (D), the suspended weight (S), and the saturated weight (W) of the sample. Suspended weight is measured for the sample suspended in water and saturated weight refers to the weight after water saturation. For detailed test specifications refer to ASTM standard C830-00. The density and apparent (open) porosity were calculated as follows.

$$V \text{ (cm}^3\text{)} = W - S \quad (1)$$

where V = exterior volume

$$\text{Volume of open pores (cm}^3\text{)} = W - D \quad (2)$$

$$\% \text{ Apparent porosity} = \left(\frac{W - D}{V} \right) \times 100 \quad (3)$$

$$\text{Bulk density} = D / V \quad (4)$$

17

Table 4 shows the calculated apparent porosity and density values for a representative sampling of MgO tapes.

3.2 SEM Analysis

SEM images of the MgO tapes prepared with and without HDPE pore formers from the 1200°C calcined MgO powder are given in Figure 8. Figure 8(a) shows the typical microstructure of MgO tapes before BBO and sintering. As-cast MgO tapes have a uniform morphology with some porosity present throughout. The densities of the green tapes produced in this study, measured as described above, were in the range of 30-44% of the theoretical density. Figure 8(c) shows the surface of the MgO tapes after BBO. At this point, the tape has been heat treated below the sintering temperature, resulting in a uniform surface with porosity throughout and with no ceramic grain structure. Figure 8(e) shows the same tape after sintering at 1400°C for 2 hours to promote grain growth and densification. The ridges seen within the MgO grains are crystallographic planes indicating grain growth during the thermal treatment. Typical densities of sintered tapes were in the range of 47-75% of theoretical density (refer to Table 4). In comparison, HDPE particles (particle size range up to ~18 μm) were added to several MgO tape casting batches to assess their effect on final tape porosity. The green MgO tapes with HDPE additions are shown in Figure 8(b) and the inset in Figure 8(b) shows the spherical, pore former particles. The resulting tapes after BBO and sintering are shown in Figures 8(d) and (f), respectively. The visible pores distributed throughout the microstructure resulted in part from the HDPE additions. HDPE is added to the tape casting slurry, remains in it during casting and

is eliminated during BBO and sintering to leave pores in the final sintered tape. Although the scope of this project did not allow detailed investigation, in principle the extent of porosity can be controlled by varying the particle size and amounts of HDPE spheres added to the tape casting slurry. This study demonstrated that the reported tape casting process can accommodate HDPE pore formers, but it did not optimize their amounts or determine quantitatively their effects on sintered tape density. The measured theoretical density of the tape shown in Figure 8(f) is 60%.

Table 4. Calculated apparent porosity and density of MgO tapes after sintering (1400°C, 2 hrs.) and creep flattening (1400°C, 4 hrs.). Samples 15-17 were prepared with 10 volume % HDPE pore former to control the final tape porosity. Sample numbers are in chronological order. The tighter limits on properties of later experiments reflect operator experience.

Sample	D Dry Weight, g	S Suspended Wt., g	W Saturated Wt., g	V Exterior Volume, cm ³	Volume of open Pores, cm ³	% Apparent Porosity	Bulk Density, g/cm ³	% Theor. Density
1	0.1560	0.1150	0.1860	0.0710	0.0300	42.25	2.20	61.03
2	0.1552	0.1069	0.1840	0.0771	0.0288	37.35	2.01	55.92
3	0.1547	0.1088	0.1912	0.0824	0.0365	44.30	1.88	52.15
4	0.1552	0.1096	0.1788	0.0692	0.0236	34.10	2.24	62.30
5	0.1608	0.1130	0.1905	0.0775	0.0297	38.32	2.07	57.63
6	0.1581	0.1120	0.1877	0.0757	0.0296	39.10	2.09	58.01
7	0.1494	0.1032	0.1755	0.0723	0.0261	36.10	2.07	57.40
8	0.1602	0.1170	0.1907	0.0737	0.0305	41.38	2.17	60.38
9	0.1640	0.1162	0.1884	0.0722	0.0244	33.80	2.27	63.10
10	0.1600	0.1128	0.1927	0.0799	0.0327	40.93	2.00	55.63
11	0.1511	0.1062	0.1774	0.0712	0.0263	36.94	2.12	58.95
12	0.1592	0.1132	0.1858	0.0726	0.0266	36.64	2.19	60.91
13	0.1502	0.1050	0.1660	0.0610	0.0158	25.90	2.46	68.40
14	0.1503	0.1067	0.1685	0.0618	0.0182	29.45	2.43	67.56
15	0.1545	0.1100	0.1717	0.0617	0.0172	27.88	2.50	69.56
16	0.1523	0.1099	0.1720	0.0621	0.0197	31.72	2.45	68.12
17	0.1591	0.1127	0.1785	0.0658	0.0194	29.48	2.42	67.16
18	0.1574	0.1114	0.1725	0.0611	0.0151	24.71	2.58	71.56
19	0.1609	0.1144	0.1791	0.0647	0.0182	28.13	2.49	69.08
20	0.1471	0.1045	0.1624	0.0579	0.0153	26.42	2.54	70.57
21	0.1444	0.1032	0.1601	0.0569	0.0157	27.59	2.54	70.49
22	0.1628	0.1155	0.1777	0.0622	0.0149	23.95	2.62	72.70
23	0.1476	0.1046	0.1617	0.0571	0.0141	24.69	2.58	71.80

3.3 Evaluation of Tape Curvature by Profilometry

Non-contact laser profilometry was used to determine MgO tape thickness, surface roughness, and overall flatness of the tape after sintering and creep flattening. For the tape casting process outlined in this report, typical MgO tape thickness and surface roughness were measured to be in the range of 200-400 μm and 10-20 μm , respectively. Figure 9 shows examples of tapes at the end of each stage of the process: (a) casting, (b) drying, (c) sintering, and (d) creep flattening.

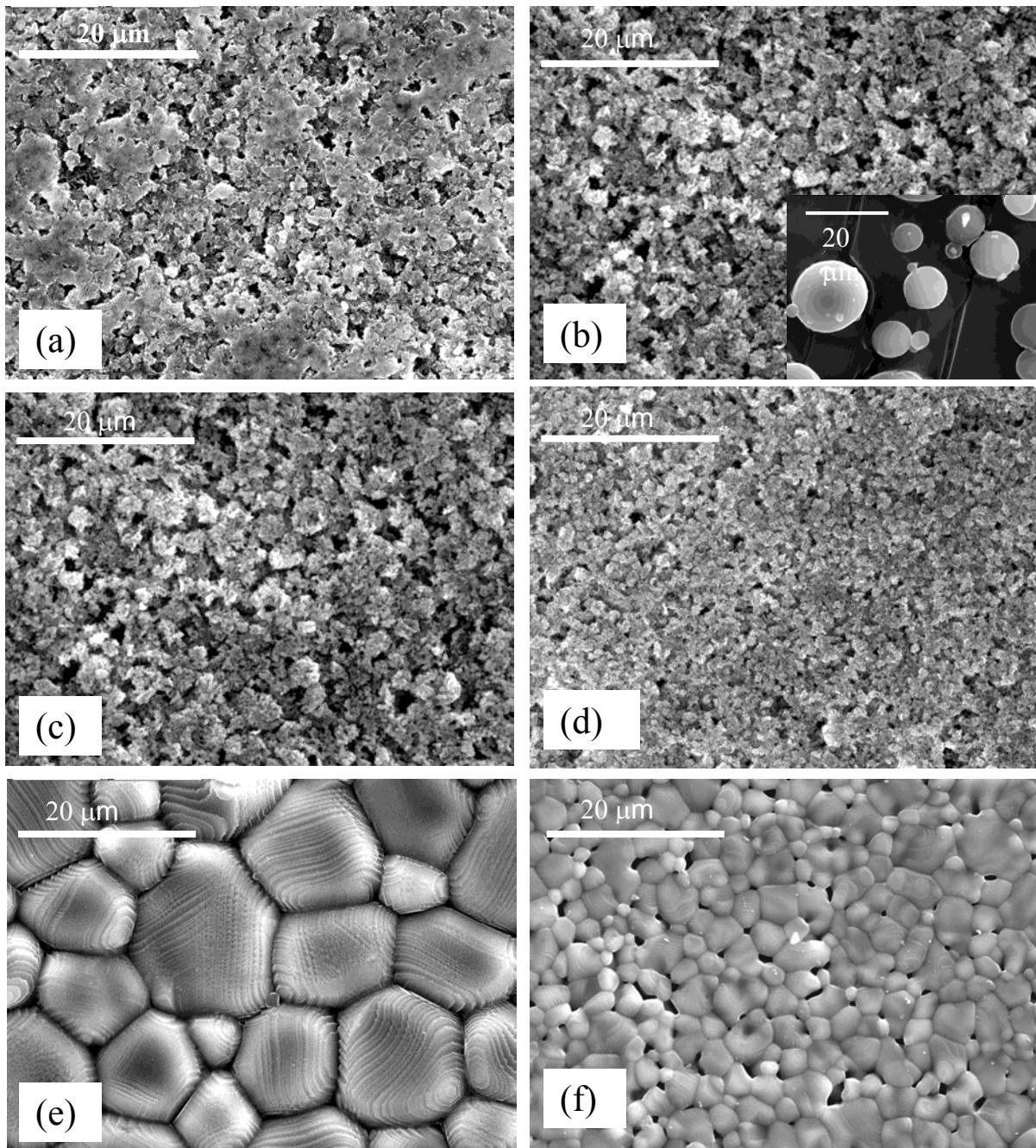


Figure 8. SEM images of MgO tapes at different processing stages. (a) Green MgO tape; (b) Green MgO tape with HDPE addition; (c) MgO tape after BBO; (d) MgO tape with HDPE after BBO; (e) MgO tape sintered at 1400°C for 2 hrs.; (f) Sintered MgO tape with controlled porosity from HDPE addition (1400°C, 2 hrs.) All images are presented at the same scale as shown in 8(a).

4. SUMMARY

A calcination study on a specified MgO powder source (Advanced Materials Inframat, LLC, Farmington, CT, USA) showed that calcining at 1200 °C for 4 hours produced a powder that was adaptable to tape casting. The resulting powder had a SSA and particle size of $14 \text{ m}^2\text{g}^{-1}$ and $1.5 \text{ }\mu\text{m}$, respectively. A tape casting procedure was developed with the calcined MgO powder to fabricate flat, sintered MgO sheets with thicknesses in the range of 200-400 μm , densities in the range of 52-73% of theoretical, and apparent porosity from 24-44%. The amount of overall linear shrinkage was measured as 20-29% and volumetric shrinkage as 48-65%.

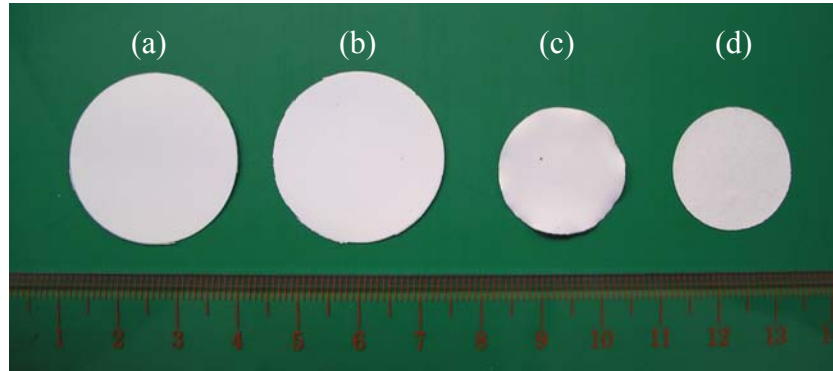


Figure 9. Digital macrographs of MgO tapes fabricated after (a) tape casting, drying and die cutting; (b) BBO; (c) sintering and; (d) creep flattening.

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