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Investigation of High Temperature Co-fired Ceramics sintering conditions using Taguchi Design of the experiment

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Abstract

Influence of sintering conditions of High Temperature Co-fired Ceramics on surface roughness, density and shrinkage of the sintered samples is presented in this paper. The investigations were conducted using Taguchi Design of the experiment. Mathematical statistics was applied in the estimation of the optimal process conditions. The estimation accuracy was verified by real measurements.

Keywords: Sintering; HTCC; Design of the experiment; Ceramics; Aqueous tape casting

1. Introduction

Low Temperature Co-fired Ceramic (LTCC) and High Temperature Co-fired Ceramic (HTCC) technologies are utilized in the fabrication of hybrid microelectronic devices [1–3]. Both LTCC and HTCC tapes are manufactured in a very similar way [4,5]. However, the tapes' content and area of applications are slightly different. High Temperature Co-fired Ceramics is more chemically and mechanically resistant, can withstand higher temperature of work and storage and has significantly (one order of magnitude) higher thermal conductivity. Therefore, in some applications HTCC technology is more recommended than LTCC. On the other hand HTCC technique is more cost consuming. After casting, the tapes are machined e.g. using punchers, lasers or milling machines, allowing various 3D mechanical structures such as fluidic channels, suspended bridges and beams to be manufactured. Afterwards, conductive paths are printed and tapes are aligned, laminated (joined) and fired. LTCC is used mainly in the fabrication of multilayer electronic substrates, wireless components/devices sensors, actuators and microsystems [1,2]. The High Temperature Ceramics is utilized in automotive applications [6], wireless communication and other RF packages [7–10] and high power application [8].

Commercial LTCC tapes are produced using tape casting based on organic solvents, whereas for HTCC, water-based tape casting has been developed during the last 20 years [11,12]. Organic solvents have lower boiling points and less energy is required in the drying step. However, they are typically toxic and flammable and, thus, not safe for the environment and users. Aqueous systems are non-toxic and inflammable. However, the slurry preparation is more complicated and more process parameters dependent, but due to efficient dispersants, high solid loadings can be reached for water-based slips. In total, an environmental assessment [13] has shown that water-based tape casting causes 30% less CO_2-emmissions than organic solvents. The aim of this paper is to investigate the influence of sintering process parameters, in terms of peak firing temperature, heating rate and dwell time at the peak firing temperature on surface roughness, shrinkage and density of sintered alumina tapes. The experiment was conducted according to statistical Design of the Experiment methodology [14–16] rules, using alumina tapes made by aqueous tape casting with properties that has previously been optimized for lamination and laser cutting [17].

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2. Design of the experiment

The use of Taguchi Design of the Experiment methodology allows reduction of the experiment time and increases the amount of information that can be obtained from the analysis. The reduction of minimal necessary combinations of investigated process parameters is met by using specially designed and optimized experiment orthogonal arrays [14]. The array which fulfills the requirements of this analysis consists of only 9 combinations of input parameters values. The arrays were created after many investigations carried out by Taguchi [14,15]. The significance of each input and the percentage influence of each of them on all analyzed output can be predicted by the analysis of variance (ANOVA) [16]. This method, thanks to mathematical statistics, permits to find the relationship between input and output precisely. Moreover, it enables a rough estimation of the results that should be achieved for a particular combination of process parameter values. In addition, the Taguchi concept permits the application of Signal to Noise (*SN*) ratio in the calculations. This solution enables to find the most optimal results from average value and deviation of average value points of view [14]. The Signal to Noise ratio presents the relation between the useful result (signal) and deviation of measured values (noise). If the deviation is high then *SN* will be lower. This method helps avoiding process conditions which increase the uncertainty of the results. Furthermore, this ratio simplifies the optimization process, because the higher *SN* the better results were achieved (more optimal), and a low *SN* indicates that the uncertainty of the result is significant and that any trends observed for results with low *SN* are just stochastic noise. Exemplary calculations using Taguchi concept are presented in Appendix A. The Taguchi concept was chosen because it was evaluated as more accurate than other methods [18–20].

3. Theory of sintering

In the firing process the ceramic particles sinter to a hard material and the sample shrinks typically 15–20%. During the sintering there is inevitably a grain growth process which generally reduces porosity and increases the density of the sample. The grain growth is enhanced by high sintering temperature and long time. Larger grains increase the long-crack toughness but decrease the strength (above approx. 5 μm) [21] which is why the grain size is generally kept as small as possible. The strength of alumina and other brittle ceramic materials is determined by the inter-grain strength. Pores and other defects decrease the contact between grains and thus decrease the strength, with larger impact the larger the defects are. The strength is limited by the largest defect in the sample [22]. There is a risk of abnormal grain growth [23], when a few grains grow much faster than the surrounding grains and act as defects in the material. When this happens pores are often trapped inside the grains and further densification is more or less stopped [24].

4. Experiment

The influence of three sintering process parameters: the peak temperature, the dwell time and the heating rate on the sintered alumina properties was analyzed in this work. The following sintered alumina properties were measured: surface roughness, shrinkage and density. The peak temperature is the highest sintering temperature and the dwell time is the time the peak temperature was held. The heating rate describes the heating rate which was used after proper debinding of tapes (above 500 °C). The heating rate for the debinding process was set to 1 °C/min. Hence, the binder was completely driven off during debinding and does not influence the experiment results.

The samples used in the study were two HTCC square tapes $20 \times 20\ mm^2$ (in green state), laminated under 20 MPa, for 10 min at 70 °C. The tapes were made by aqueous tape casting using alumina powder (AKP30, Sumitomo Chemical), dispersant (Dolapix PC21, Zschimmer & Schwarz) and latex binder (50/50 of Resicel E50N, Lamberti Speciality Chemicals, and LDM 7651S, Celanese Emulsions). The binder composition was previously optimized for lamination and laser cutting [17].

The experiment was conducted according to statistical Design of the Experiment methodology [14–16] rules. If three process parameters are investigated in the analysis, then, according to Taguchi [14], an orthogonal array with only 9 combinations of input parameter values can be utilized. The examined values for all three investigated sintering process parameters are given in Table 1, and the 9 combinations of input parameter values in Table 2. The cardinality of samples for each combination was 9, hence the total number of samples was 81. However, the shrinkage was measured in 4 places of each sample; hence the total cardinality for this part was 324. The surface roughness was measured from both sides of all substrates and therefore, the cardinality for this analysis was 162.

Table 1
Analyzed sintering process parameters.

Parameter	Parameter name	Acronym	Parameter value		
			***Lower level* (1)**	***Nominal level* (2)**	***Higher level* (3)**
P1	Peak temperature (°C)	P1	1550	1575	1600
P2	Dwell (min)	P2	5	50	90
P3	Heating rate (°C/min)	P3	1	4	7