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Comparison of Lamination Methods to Control Low Temperature Co-fired Ceramic Shrinkage

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Abstract

The process of low temperature cofired ceramics fabrication includes a lamination step where pressure is applied during a certain time and at a certain temperature in order to create a monolithic block out of the green sheets before firing. This lamination can be done by the use of a uniaxial or an isostatic press. In this paper, we compare the two methods to determine if there is a difference in lamination quality and if the shrinkage after firing is impacted by the lamination method. The study is based on a design of experiments approach, where the four factors – stack thickness, surface area, lamination pressure and lamination temperature – are tested using an isostatic press. The result variables are lamination quality, vertical and lateral shrinkage. The findings are compared to a previous study where the same set of experiments was performed with a uniaxial press. It is found that the isostatic press allows for a better lamination quality for small surface area, high stack samples. Also, the isostatic lamination leads to less shrinkage variation. Finally, we present prediction models for the vertical and lateral shrinkage, that will allow the manufacturer to better predict and control the fabrication process.

Key words

Lamination, uniaxial, isostatic, DOE, shrinkage.

I. Introduction

Low Temperature Cofired Ceramics (LTCC) is normally produced according to a standardized scheme as given by tape manufacturers. A typical process overview is given in Fig. 1.

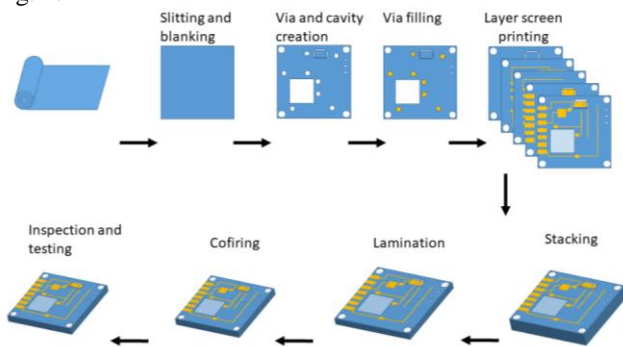


Fig. 1. LTCC process overview

However, each manufacturer has to adapt this general scheme to his equipment in order to obtain a satisfactory level of repeatability. One of the well-known difficulties of LTCC is the control of the shrinkage that occurs during firing

as the organic slurry outgasses allowing the LTCC particles to densify into a homogenous block. One of the manufacturing steps is the lamination which is normally given by the tape provider to be performed by applying 21 MPa pressure for ten minutes at +70°C. Some LTCC users operate with an isostatic press for this lamination while other function with a uniaxial press. During a first work [1], we have tested the impact of the parameters stack thickness, surface area, applied pressure and lamination temperature using a uniaxial press by the setup of a Design of Experiment (DOE) [2]. In this first study, it was concluded that the lamination quality is mainly controlled by the interaction between the stack thickness and the surface area. For the z-direction shrinkage the stack thickness and surface area interaction together with lamination temperature were found to be significant factors. Predictions models were elaborated for vertical and lateral shrinkage. The lateral shrinkage is mainly impacted by the two main effects stack thickness and surface area while the temperature has a smaller impact and the applied pressure is non-significant in the tested range. In this paper we investigate the influence of the same parameters in an isostatic lamination process, in order to

understand if the isostatic press can provide advantageous results as compared to the uniaxial press.

II. Design of Experiment

In order to perform a fair comparison, the same DOE setup as in [1] is prepared with 10 mil thick L8 tape from Ferro [3] using an isostatic press IL-4004 from PTC, to test the impact of four parameters: surface size, stack thickness, lamination pressure and lamination temperature, with levels as indicated in TABLE I.

TABLE I. EXPERIMENTAL FACTORS

Factor	Unit	Symbol	Minimum value	Maximum value
Stack thickness	μm	A	508	2032
Surface area	mm^2	B	25.4×25.4	50.8×50.8
Applied pressure	MPa	C	9 ± 0.5	17 ± 0.5
Temperature	$^{\circ}\text{C}$	D	40	70

In this way, the least possible bias is introduced. The samples are prepared as in [1], i.e. the tapes are cut, the alignment holes are laser cut, the top layers are screen printed with FX30-025JH gold paste and the stacking is done at IMT Atlantique before placing each sample in a sealed vacuum bag to be sent off to Landshut University for isostatic lamination. At Landshut, the stacks were taken out of their individual bags, Teflon inserts were placed in the alignment holes (to avoid that the lamination bags would tear during lamination), the stacks were placed in lamination bags with a lamination foil underneath and on top and a stabilizing alumina substrate at the bottom of the stack with the purpose to preserve the flat shape during lamination. Since the isostatic lamination chamber is large enough to place several bags within the chamber simultaneously, the samples were laminated two by two to save time. That is, the samples with the same lamination pressure and duration were laminated simultaneously (e.g. samples 1 and 2). TABLE II describes the fixed factors and TABLE III the DOE setup.

TABLE II. FIXED FACTORS

Factor	Fixed value	Comment
Operator		One operator at IMT Atlantique and one operator at Landshut
Tape material	Ferro L8-10	Each layer is 10 mil (254 μm) thick before lamination
Holes/cavities/vias	None	Except for stack alignment holes
Screen printed paste	FX30-025JH	On top layer only
Press	Isostatic	PTC IL-4004
Lamination time	5 minutes	
Simultaneous lamination	Yes	Two sample pressed each time (same pressure and temperature)

Sintering furnace	Nabertherm L9	
Simultaneous sintering	Yes	All samples sintered at the same time
Sintering	850°C , 30 minutes	Firing profile according to manufacturer's data [3]

TABLE III. 2^{4-1} DOE EXPERIMENT. THE -/+ SIGNS IN THE ABC AND D COLUMNS, INDICATE THAT THE MINIMUM OR THE MAXIMUM VALUE IS USED, RESPECTIVELY, 0 INDICATES THE USE OF THE CENTRE VALUE.

Sample	A	B	C	D	AB	AC	BC
Alias structure	BCD	ACD	ABD	ABC	CD	BD	AD
1	-	-	-	-	+	+	+
2	+	+	-	-	+	+	-
3	+	-	+	-	-	-	-
4	-	+	+	-	-	-	+
5	-	-	+	+	+	+	-
6	+	+	+	+	+	+	+
7	+	-	-	+	-	-	+
8	-	+	-	+	-	-	-
9	0	-	0	0			
10	0	+	0	0			

A. Experimental settings

Due to tolerances in control of the isostatic laminator the actual process parameters can differ from the chosen DOE values. First, the pressure in the lamination chamber is built up within several (~10) pumping cycles with a relatively primitive control and is indicated by a rather coarse pressure gauge. The number of pumping cycles can vary, which is not considered in time control of the process duration, so the effective lamination duration can also slightly change. Additionally, the water temperature in the lamination chamber has a gradient of up to 5°C from the front to the backside while the thermoelement is placed in the center of the tank, thus the precision of the temperature control is limited. TABLE IV presents the DOE parameter values as they were measured in the experiments.

TABLE IV. MEASURED PARAMETER VALUES

Sample	Pressure [MPa]	Temperature [$^{\circ}\text{C}$]	Duration [sec]
1, 2	9.3	41	300
3, 4	16.8	40	300
5, 6	17.3	70	300
7, 8	9.0-8.5	68	330
9, 10	13	55	300

III. Results

The results variables are lamination quality, vertical and lateral shrinkage. The lamination quality is judged perfect if there is no identifiable gap between the layers after cross sectioning of the samples. Thus, the gap size in μm is retained as the result variable. The vertical (z-) shrinkage is calculated as the after-firing stack height compared to stack height before lamination. This might then differ from the manufacturers z-shrinkage which is in most cases calculated as the shrinkage obtained after firing compared to the stack

height measured after lamination. Finally, for the lateral x- and y- shrinkages, we evaluate the distance between external edges of the screen-printed squared bars pattern after firing as compared to the designed value, in percent. The x- and y- shrinkage is given individually for all samples, but the model for future use is calculated from the x- and y- mean value to give the lateral shrinkage of each sample. Two of the produced samples after firing are shown in Fig. 2.

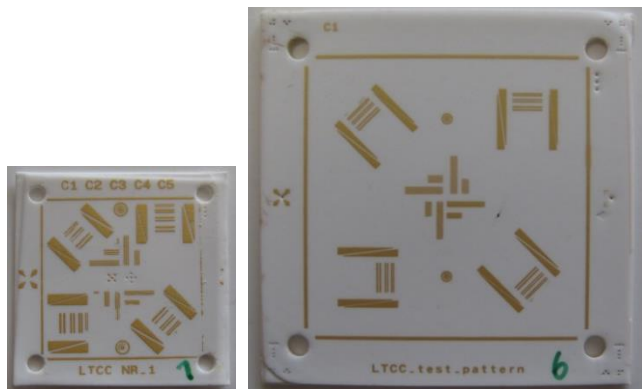


Fig. 2. Sample number 1 (-,-,-,-) and sample number 6 (+,+,+,+) after firing.

The evaluation of experimental results is presented in TABLE V.

TABLE V. GAP SIZE AND SHRINKAGE IN X-, Y- AND Z-DIRECTION FOR THE EIGHT SAMPLES.

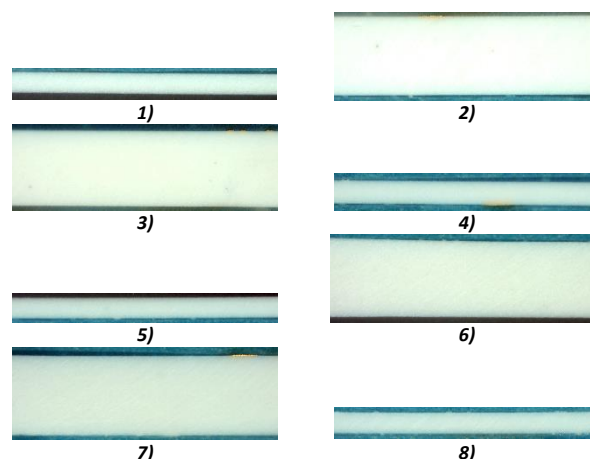
Sample	Gap [μm]	Shrinkage [%]		
		x	y	z
1	0	16.36	15.73	28.15
2	0	15.57	15.46	25.98
3	0	16.20	16.25	28.64
4	0	15.51	16.17	21.46
5	0	13.93	14.19	30.91
6	0	13.54	13.60	28.64
7	0	15.30	14.91	28.20
8	0	14.66	14.63	25.59

A. Lamination quality

The lamination quality is analyzed after cross sectioning of the samples. In our previous DOE [1], where a uniaxial press was used, samples number 3 and 7 were badly laminated, whereas all other samples were flawless. Here, using the isostatic press, all samples are well laminated, as can be seen in TABLE V and TABLE VI.

This is an interesting result, indicating the advantage of isostatic lamination, especially when small sized samples with high stack thickness are laminated.

TABLE VI. CROSS SECTIONS OF THE ISOSTATICALLY LAMINATED SAMPLES



B. Vertical shrinkage

The Pareto plot is presented in Fig. 3. As can be seen, none of the tested factors has a significant effect on the result variable, whereas in [1] the interaction AD (stack thickness and temperature) as well as factors D (temperature) and B (surface area) were judged significant.

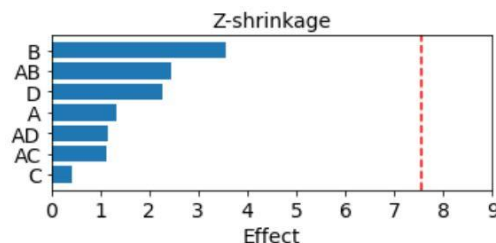


Fig. 3. Pareto plot of the effects on the vertical shrinkage result variable.

In this case, even though judged insignificant, the largest effect is obtained for the surface area (B), followed by the interaction between stack thickness and surface area (AB) and temperature (D). The applied pressure has the smallest effect of all factors and interactions.

In Fig. 4 the main effect plots of the vertical shrinkage are presented. It can be seen that smaller stack thickness (A), larger surface area (B) and lower temperature (D) result in less shrinkage. For the applied pressure (C) we get an almost horizontal line, so this factor has a very limited effect on the result.

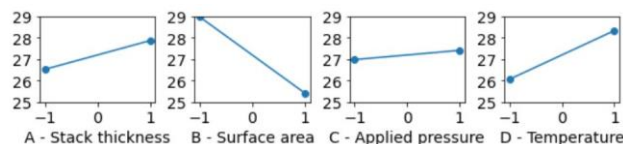


Fig. 4. Main effect plot of vertical shrinkage.

The interaction plots in Fig. 5 show that for a low stack thickness a small surface results in an increased shrinkage,

while for a large stack thickness the surface area impact is less pronounced. The interaction between stack thickness and pressure is valid (since the two lines crosses each other) with a more important pressure induced change if the stack thickness is high. Finally, a low stack thickness induces a higher impact of the temperature than does a high one.

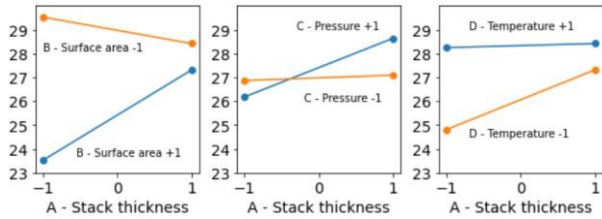


Fig. 5. Interaction plots for vertical shrinkage.

From these results, a regression model where A, B, C and D stands for the factors' normalized values ranging from -1 to +1 is produced (1). The mean vertical shrinkage is found to be 27.20 %.

$$Z_{shrinkage} = 27.20 + 0.6704A - 1.778B - 0.2154C + 1.138D + 1.224AB + 0.5559AC - 0.5844AD \quad (1)$$

C. Lateral shrinkage

To simplify the lateral shrinkage analysis and to prepare for use in design work, we have evaluated the x- and y-shrinkages for each sample as a combined lateral shrinkage. Thus, the two individual results for each setting are seen as a repetition. Having done this, the resulting Pareto plot is presented in Fig. 6 and the main effect plots in Fig. 7

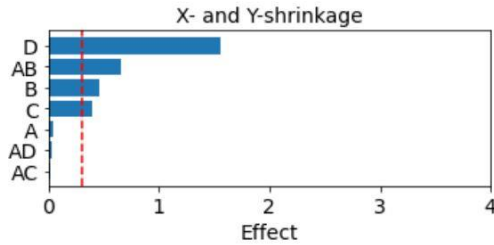


Fig. 6. Pareto plot of the lateral shrinkage for isostatic lamination.

From these results we have four significant factors, the lamination temperature (D), followed by the interaction between stack thickness and surface area (AB), then comes the surface area (B) and pressure (C). This is once again different from the results using a uniaxial press where stack thickness (A), surface area (B) and temperature (D) were significant factors.

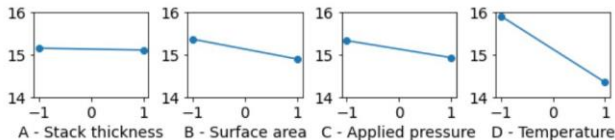


Fig. 7. Main effect plots of the lateral shrinkage

Looking at the main effect plots in Fig. 7, we clearly see that decreasing any of the factors from the standard values, will result in an increased shrinkage. The interaction plots in Fig. 8 show that merely interaction between the stack thickness and the surface area exists, since in the two other plots the lines are parallel and never cross. If the stack thickness is at its low value (-1) the surface area has little impact on the result, but with a high stack thickness a smaller surface area will result in a higher shrinkage. This is the opposite of what was found for the vertical shrinkage.

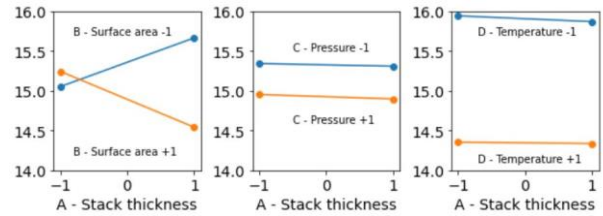


Fig. 8. Interaction plots for lateral shrinkage.

The regression model for isostatic lamination is found to be as in (2). The mean lateral shrinkage of the L8 tape is 15.1 % whereas for the uniaxial press it was 13.3 %. Hence, we have a stronger lateral and a weaker vertical shrinkage when using an isostatic press compared to a uniaxial one.

$$Lateral\ shrinkage = 15.1261 - 0.223A - 0.2331B - 0.2004C - 0.7808D - 0.3280AB - 0.0057AC + 0.0141AD \quad (2)$$

D. Result validation

To test the two regression models, we added two samples with midpoint values for all factors except the surface area. The predicted shrinkages can then be compared with the calculated ones. Thus, we used 1270 μm stack thickness, 25.4 mm × 25.4 mm surface area, 13 MPa and 55°C for the number 9 sample and 1270 μm stack thickness, 50.8 mm × 50.8 mm surface area, 13 MPa and 55°C for sample number 10.

The cross sections in TABLE VII. show, as foreseeable, excellent lamination quality with no delamination.

TABLE VII. CROSS SECTION OF THE VALIDATION SAMPLES

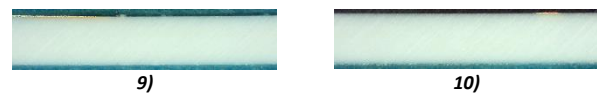


Fig. 9. Cross sections of sample 9 and sample 10. The lamination quality is excellent.

In TABLE VIII and TABLE IX, we present the expected and the obtained results, respectively.

TABLE VIII. ACCORDING TO REGRESSION MODELS

Run	A	B	C	D	Lamina- tion gap [μm]	Shrinkage [%]		
						x	y	z
9	0	-	0	0	0	15.36	15.36	28.98
10	0	+	0	0	0	14.90	14.90	25.42

TABLE IX. OBTAINED RESULTS

Run	A	B	C	D	Lamina- tion gap [μm]	Shrinkage [%]		
						x	y	z
9	0	-	0	0	0	14.46	14.54	28.98
10	0	+	0	0	0	14.31	14.43	26.77

The obtained shrinkage is within a one percent unit margin for the lateral shrinkage and is slightly larger for the vertical shrinkage. This can be considered as a sufficiently good result as the regression models are based on idealized settings and not on the real parameter values, as discussed earlier in this paper.

IV. Comparison of Uniaxial and Isostatic Lamination

In order to be able to conclude on the difference between the isostatic and uniaxial lamination we need to compare the obtained results. TABLE X summarizes our findings.

TABLE X. COMPARISON BETWEEN UNIAXIAL AND ISOSTATIC LAMINATION RESULTS

	Uniaxial	Isostatic
Lamination quality	Not OK for A+ B- case	OK for the whole parameter range
Z-shrinkage		
Significant factors	AB, D, B	None (B, AB, D largest effects)
Regression model	$32.28 - 0.04625A - 0.7262B - 0.4562C + 1.919D - 2.351AB - 0.2112AC + 0.09875AD$	$27.20 + 0.6704A - 1.778B - 0.2154C + 1.138D + 1.224AB + 0.5559AC - 0.5844AD$
Mean shrinkage value and standard deviation	$32.3 \pm 3.2\%$	$27.2 \pm 2.7\%$
XY-shrinkage		
Significant factors	A, B, D	D, AB, B, C
Regression model	$13.319 - 1.648A + 1.491B - 0.197C - 0.366D - 0.192AB - 0.059AC - 0.068AD$	$15.1261 - 0.223A - 0.2331B - 0.2004C - 0.7808D - 0.3289AB - 0.0057AC + 0.0141AD$
Mean shrinkage value and standard deviation	$13.3 \pm 2.3\%$	$15.1 \pm 0.9\%$

The isostatic lamination works as intended for the whole proposed range of parameters, while for the uniaxial

lamination high stack thickness (A) in combination with small surface area (B) resulted in failed lamination. In this study, we found no significant factors for the vertical shrinkage even though the same factors as in the uniaxial case had the largest effect although in a different order. The vertical shrinkage mean value amounts to 27.2 % while it was 32.3 % in the uniaxial case. The only significant factor was found to be the temperature (D) while in the uniaxial case we had three significant factors: the stack thickness (A), the surface area (B) and the temperature (D). In both cases, the pressure (C) was not significant. The lateral shrinkage mean value for the isostatic case is found to be 15.1 % while it was only 13.3 % in the uniaxial case. Thus, we observe lower vertical and higher horizontal shrinkage in case of isostatic lamination. The standard deviation of the shrinkage has a higher value in the uniaxial case, especially in the lateral direction. The lower variation is off course advantageous when it comes to fabrication of LTCC devices.

V. Conclusion

Comparing isostatic and uniaxial lamination of LTCC we have found substantial difference. In the uniaxial case it is difficult to produce small surfaced high stacked devices. The isostatic lamination leads to less shrinkage variation and can be controlled by one most significant factor – the temperature. This is true even though the parameters of the lamination process could not be perfectly controlled. In both cases, models are proposed to help manufacturers predict the shrinkage and adjust their designs accordingly.

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