Control of Low Temperature Co-fired Ceramic Shrinkage for Unconstrained Sintering

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Abstract— The shrinkage of Low Temperature Co-fired Ceramics (LTCC) during firing is one of the most difficult features to control in LTCC fabrication, as many factors may impact on the result. The shrinkage given by the tape manufacturer is not perfectly transposable to a production environment where preparation, use and equipment is not in exact accordance. Thus, predictable shrinkage models are of main importance in order to fabricate LTCC devices according to specifications. The objective of this work is to develop such models for the Ferro L8 tape using the powerful Design of Experiments (DOE) technique. Four factors are varied; the stack thickness, the device surface, the applied pressure and the temperature during lamination. Other factors such as operator, lamination time or firing profile are kept to a fixed value during these experiments. The result variables are lamination quality and x, y and z-direction shrinkage. Lamination quality is found to be mainly impacted by the interaction between the stack thickness and the surface area of the stack, while for the z-direction shrinkage this interaction together with lamination temperature are significant factors and finally for the lateral shrinkage the main effects stack thickness, surface area and temperature are significant. Numerical models for shrinkage in z- and lateral directions are established. This work enforces the understanding of the shrinkage of LTCC and permits for the Ferro L8 users correctly compensate the layout for shrinkage.

Keywords—Low Temperature Co-fired Ceramics (LTCC); shrinkage; lamination; Design of Experiments (DOE);

I. Introduction

The manufacturing steps for Low Temperature Co-fired Ceramics (LTCC) are generally as follows: cutting tape's outer dimensions, cutting or punching holes and cavities, via hole filling, screen printing, stacking, lamination and firing. During firing the substrate shrinks due to outgassing of volatile material and densification of the ceramic powder that is present in the tapes. The shrinkage is often defined by the LTCC tape manufacturer as the dimensional difference in lateral (x- and y-) and vertical (z-) direction after firing, compared to the dimensions obtained after 10 minutes lamination at +70°C with 21 MPa of applied pressure. These hot lamination settings are rarely explained, little information on their impact of the resulting LTCC device is given as compared to cold chemical lamination which is well represented in literature. The lamination parameters given in early LTCC related patents are also very limited, for instance [1] and [2] mentions lamination without giving any processing data. In [3] some data regarding the lamination density vs x- and y-shrinkage is given, although without any further information to the initial choice and importance of temperature and time.

If the lamination parameters are different than what is given by the tape manufacturer, this may impact the lamination quality as well as the shrinkage. The same holds for the very nature of the circuit i.e. tape material, number of layers, x- and y-dimensions, number of filled via holes, cavities, metal paste or if any other paste is used (resistive or dielectric), as well as the in-house procedure and equipment used to accomplish the circuit. This is why LTCC tape users should do tests to find their proper shrinkage and lamination quality outcome based on their production facility and product design. One way to do this in a controlled way is to use the technique of design of experiments (DOE), [4].

DOE is a powerful tool to test the impact of different factors on the result variables. It also allows for a development of predictive models to anticipate future results, as long as one uses parameter settings within the tested minimum and maximum range values of each tested factor. A full factorial DOE design requires a minimum of 2^N samples, where N is the number of factors to be tested. If N is large, screening methods exist to reduce the number of required samples.

In [5] a DOE using the DuPont 951AX tape was performed varying temperature, lamination duration, lamination pressure and number of layers in order to propose a model for prediction of shrinkage. The significant factors in this work were found to be the lamination pressure and lamination temperature while number of layers and duration of lamination could be neglected.

In [6] metal loading, lamination pressure and layer count were taken into consideration, while lamination time and temperature were fixed values. Here, the Dupont 951P2 tape was used and it was found that the metal loading and lamination pressure were significant factors. These factors are represented in the shrinkage model that resulted from this work.

Based on these two papers and on our own experience we have initiated a DOE with the goal to obtain a model that predicts the parameter settings needed to achieve a certain shrinkage for the Ferro L8 LTCC material [3]. According to the manufacturer, the x-and y-direction shrinkage is typically 13.3 ± 0.3 %, and z-shrinkage is typically 30 %, when standard lamination procedure and settings are used.

II. Experiment Design

Factors that could impact the lamination results are the LTCC material, the number of layers (judged non-significant [5]-[6]), the layers' thickness, the circuits surface area, placement of holes, metallized vias, cavities and pattern, screen printing paste, the applied lamination pressure, the temperature during lamination and the time of lamination (also discarded from [5]). From [5], lamination pressure and temperature and from [6], lamination pressure and metal loading were found significant, out of their tested factors.

Other factors such as room temperature and humidity, age of tape, operator, press type (uniaxial or isostatic), vacuum treatment of stack before lamination, number of screen-printed layers, etc. may also have an impact, but as we cannot treat them all, we'll stabilize these factors to have as little variation as possible.

A. 2^{4-1} DOE design

In this work we have decided to concentrate on the following four factors: the stack thickness (which combines the layer thickness and number of layers into one factor), the LTCC circuits size (surface area), the applied pressure during lamination and the lamination temperature. We'll use one single LTCC material, the Ferro L8 tape from the same production batch, we'll have no holes (except the stack alignment holes), metallized vias or cavities and the lamination time will be set to 5 minutes for all of the tests. Also, one single operator will perform all the fabrication steps of the samples the same day and they will all be fired simultaneously in the static programmable furnace to avoid as much variation as possible. The holes, filled vias, cavities and other tape materials are factors that may be tested in further work.

TABLE I. below presents the chosen factors with their minimum and maximum settings. The pressure range is set from 9 MPa to 17 MPa, which is not conformal to the proposed lamination pressure given by the LTCC tape manufacturer [3]. This range is chosen to fit to the inhouse uniaxial press, which is a simple benchtop press from ColorKing. TABLE II. lists the non-varied factors and their settings throughout this experiment.

TABLE I. EXPERIMENTAL FACTORS

Factor	Unit	Symbol	Minimum value	Maximum value
Stack thickness	µm	A	508	2032
Surface area	mm²	B	25.4 × 25.4	50.8 × 50.8
Applied pressure	MPa	C	9±0.5	17±0.5
Temperature	°C	D	40	70

TABLE II. FIXED FACTORS

Factor	Fixed value	Comment
Operator		The same for all operations²
Tape material	Ferro L8-10	Each layer is 10 mil (245 µm) thick before lamination
Holes/cavities/vias	None	Except for stack alignment holes
Screen printed paste	FX30-025JH	On top layer only
Press	Uniaxial	Colorking AR1706
Lamination time	5 minutes	
Simultaneous lamination	No	One sample pressed at a time
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]

A full factorial design would require 1290 cm² of tape which is a considerable amount, therefore we chose to do a fractional factorial experiment of 2^{4-1}, i.e. eight samples which necessitates half the amount of tape. By this half-fractional design, we will sacrifice a third-order interaction to generate the settings of one of the initial four factors.

The DOE 2^{4-1} experiment is performed according to the settings in TABLE III. columns A, B, C and D. The interactions are also presented in this table as well as the aliasing structure that comes from the fractional design. The run order is randomized to avoid any bias due to humidity or ambient temperature deviation, to the machines heating up or any other time-dependent factor.

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

Run order	A	B	C	D	AB	AC	BC
Alias structure	***BCD***	***ACD***	***ABD***	***ABC***	***CD***	***BD***	***AD***
1	-	-	-	-	+	+	+
7	+	-	-	+	-	-	+
8	-	+	-	+	-	-	-
2	+	+	-	-	+	+	-
5	-	-	+	+	+	+	-
3	+	-	+	-	-	-	-
4	-	+	+	-	-	-	+
6	+	+	+	+	+	+	+

B. Result variables

The lamination quality is the result variable of principal interest. If we do not achieve a good lamination, an improved knowledge of shrinkage for the used settings is useless. To measure the lamination quality, we will do cross sections of the eight substrates and measure the gaps between layers.

Once the lamination is correct the anticipated result variables of shrinkage in x- and y-directions are relevant. A model for these values will help the user to anticipate the shrinkage during his preparation of the fabrication.

III. EXPERIMENTAL RESULTS

In Fig. 1, two of the eight test samples after lamination, are presented. The gold pattern is only screen printed on the top layer. Its outer square pattern is used to measure the shrinkage in x- and y-directions. The x- and y-shrinkage values are calculated as the difference in percent after firing as compared to the value after lamination, while the z-shrinkage is calculated as the difference in percent after firing as compared to the green tape stack thickness before lamination. The magenta colored deformation on each substrate edge is caused by nail varnish that is used to fix the layers one to another before removing the stack from the stacking fixture. This is needed since the lamination is performed without the fixture. The samples after firing are presented in Fig. 2.

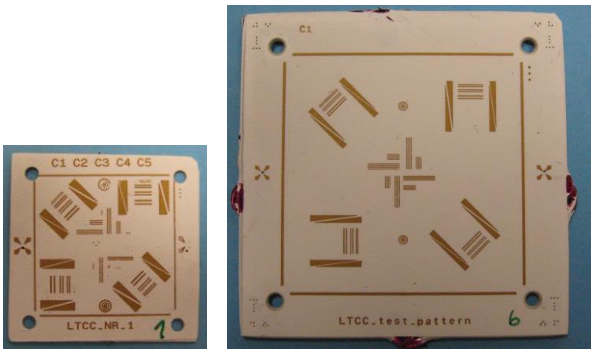


Fig. 1. Sample number 1 and 6 after lamination. The sample number equals run order number given in TABLE III.

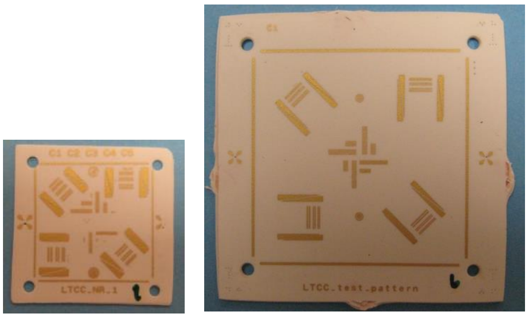
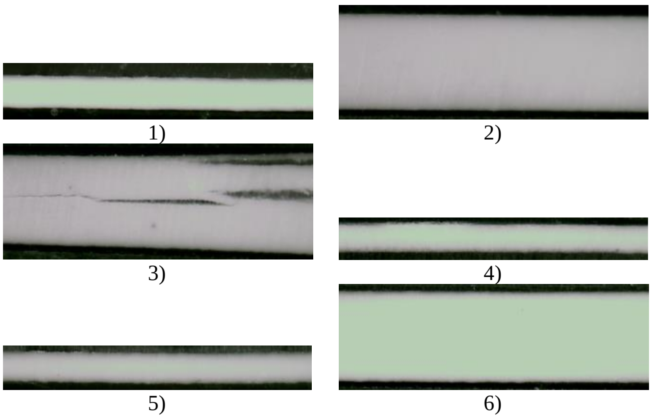


Fig. 2. Sample number 1 and 6 after firing.

The obtained results from this DOE are presented in TABLE IV. and the cross sections of each samples are presented in Fig. 3. The cross sectioning reveals that all samples but number 3 and 7 were correctly laminated.

TABLE IV. 2^{4-1} DOE EXPERIMENT RESULTS. A, B, C AND D STANDS FOR STACK THICKNESS, SURFACE, APPLIED PRESSURE AND TEMPERATURE RESPECTIVELY.

Run	A	B	C	D	Lamination gap [µm]	Shrinkage [%]		
						x	y	z
1	-	-	-	-	0	13.98	13.46	29.13
7	+	-	-	+	>100	9.62	10.77	38.00
8	-	+	-	+	0	16.49	16.49	36.02
2	+	+	-	-	0	13.43	13.89	27.81
5	-	-	+	+	0	12.98	12.72	32.28
3	+	-	+	-	>100	9.97	11.13	32.63
4	-	+	+	-	0	16.94	16.68	31.89
6	+	+	+	+	0	12.08	12.48	30.51



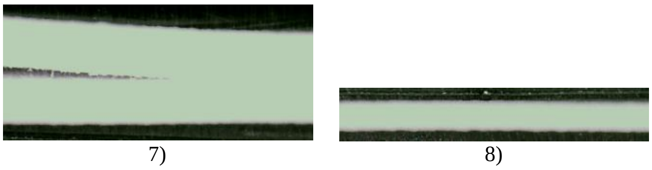


Fig. 3. Cross sections of the eight samples. Sample numbers which are equal to the run numbers are given under each micrograph.

From the results obtained as presented in in TABLE IV. , we calculate the effects and present them in Pareto plots, one for each result variable, i.e. x-y shrinkage, z-shrinkage and lamination gap, Fig. 4.

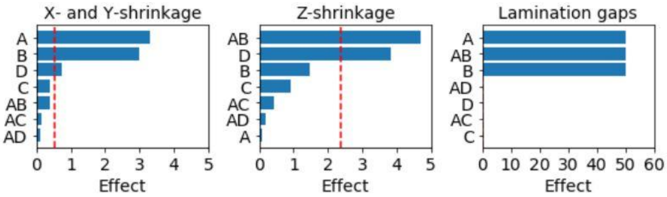


Fig. 4. Pareto plots for the three result variables.

A. Lamination gaps

From the Pareto plot of the lamination gaps we find that the stack thickness (A) and surface area (B) and the interaction between these two effects are the only significant effects. The main effect plots in Fig. 5 and the interaction plot in Fig. 6 proves the same thing. From the interaction plot of the stack thickness combined with the surface are (interaction AB) we can conclude that only when the surface area is small, the stack thickness impacts on the lamination results. Bad lamination appears when we have a high stack thickness and a small surface area.

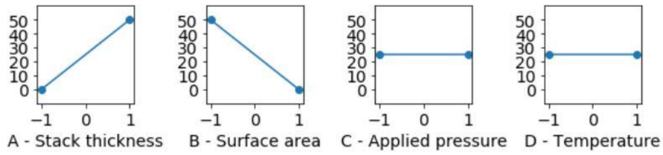


Fig. 5. Main effect plots of the lamination gap result variable.

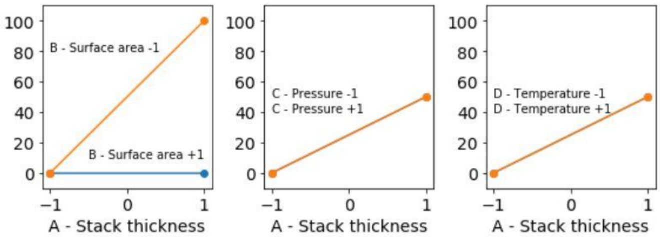


Fig. 6. Interaction plots for lamination gaps.

B. Vertical shrinkage

The z-shrinkage Pareto plot is also presented in Fig. 4. Using a 95 % confidence level, the factors that impacts the vertical shrinkage significantly are the interaction between stack thickness and surface area (AB), followed by the temperature (D). The other main effects and interactions are non-significant.

Main effect and interaction plots are presented below, Fig. 7 and Fig. 8. The stack thickness (A) has no impact of the shrinkage when looked at alone as a main effect. The surface area (B) gives higher shrinkage for less area, as does the applied pressure (C). The only main effect that impacts significantly on the z-shrinkage is the temperature (D). A higher temperature will increase the shrinkage of the LTCC stack. Further on, looking at the interactions, there is a clear interaction between the stack thickness (A) and surface area (B), which will change the outcome according to settings, see Fig. 8.

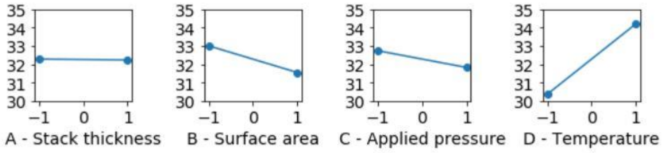


Fig. 7. Main effect plots for z-shrinkage.

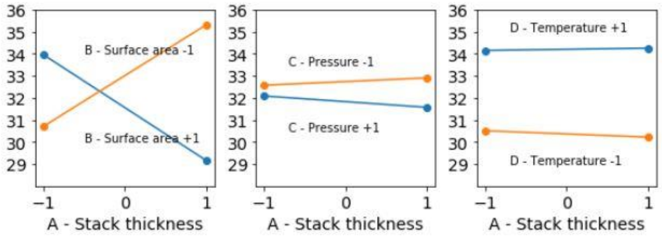


Fig. 8. Interaction plots for z-shrinkage.

1) Z-shrinkage regression model

The regression model given for this vertical shrinkage is presented in (1) where A, B, C and D stands for the factors' normalized values ranging from -1 to +1.

$$Z_{shrinkage} = 32.28 - 0.04625A - 0.7262B - 0.4562C + 1.919D - 2.351AB - 0.2112AC + 0.09875AD \quad (1)$$

The mean value is 32.28 % which is close to the value proposed by the manufacturer.

C. Lateral shrinkage

The x- and y-shrinkages as given in TABLE IV. are evaluated together as lateral shrinkage. Thus, the two individual results for each setting are seen as a repetition. This way we will be able to decide on one predictive model which is more adequate to compensate for while designing the circuits.

From the adequate Pareto plot in Fig. 4 we conclude that the stack thickness (A), surface area (B) and temperature (D) are judged significant with a 95 % confidence level, while the pressure (C) and the interactions are non-significant.

This result is contradictory to what was found in [5] and [6] and rather surprising as one would intuitively consider pressure and temperature to be factors of importance, while the surface area should have no impact nor the thickness of the stack. Essentially, the press should apply a force per area that corresponds to the set pressure. However, since we use no fixture in the press, the tape's viscosity may impact the lamination.

Main effect plots and interaction plots for the lateral shrinkage are presented in Fig. 9 and Fig. 10 respectively. The main effect plots show that more x- and y-shrinkage will result if the stack thickness is small and if the surface area is large.

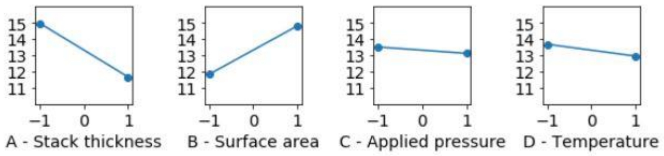


Fig. 9. Main effect plots for the combined x and y-direction shrinkage.

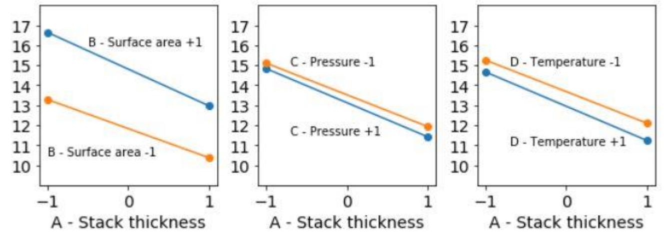


Fig. 10. Interaction plots for the combined x- and y-direction shrinkage.

There are no lines crossing in the interaction plots, thus there are no interactions between these factors.

1) Lateral shrinkage model

The regression model that come out of this experiment for the lateral shrinkage is given in (2):

$$Lateral\ shrinkage = 13.319 - 1.648A + 1.491B - 0.197C - 0.366D - 0.192AB - 0.059AC - 0.068AD \quad (2)$$

The mean value of all results for lateral shrinkage is found to be 13.319 % which is in accordance with the shrinkage proposed by the manufacturer. However, in our case, opposite to what is proposed by the manufacturer, the pressure is not a significant factor, c.f. Fig. 4.

Models (1) and (2) can now be used for all LTCC designs using the Ferro L8 tape, 25.4 mm × 25.4 mm to 50.8 mm × 50.8 mm, with a thickness from 508 to 2032 µm.

D. Result validation

To confirm this regression models, two test samples are realized, one 25.4 mm × 25.4 mm and one 50.8 mm × 50.8 mm sample, both having a thickness of 1270 µm which is the center value of this parameter, both being screen printed with the same motives and paste as the initial samples and both being laminated at 13 MPa at 55°C. All fixed factors are kept the same as before (except the fact that the firing date is not the same).

From the regression models (1) and (2) we should find the following results, see TABLE V. while the obtained results are presented in TABLE VI.

TABLE V. EXPECTED RESULTS ACCORDING TO REGRESSION MODELS

Run	A	B	C	D	Lamination gap [µm]	shrinkage		
						x	y	z
9	0	-	0	0	0	11.82	11.82	33.00
10	0	+	0	0	0	14.81	14.81	31.55

TABLE VI. OBTAINED RESULTS

Run	A	B	C	D	Lamination gap [µm]	shrinkage		
						x	y	z
9	0	-	0	0	0	10.80	10.45	35.7
10	0	+	0	0	0	13.40	14.46	31.4

The cross sections in Fig. 11 show excellent lamination quality with no delamination.

Fig. 11. Cross sections of sample 9 and sample 10. The lamination quality is excellent.

Comparing the obtained values from TABLE VI. with the expected values in TABLE V. there is a small difference. Yet, the responses are correct in the way that the smallest lateral and highest z-shrinkage results where expected, i.e. in sample 9 and the largest lateral and smallest z-shrinkage where expected, sample 10. A tolerance level should be added to the lateral shrinkage model. Since we only used one sample for each setting, the used data includes no variation, which would be the case for a repetition of the settings in the DOE.

IV. DISCUSSION

In order to fully benefit from these findings, one needs to take the three result variables into simultaneous consideration. First, in order to guarantee a good lamination quality, small surface area devises should be avoided, i.e. the factor B should be set to its high value. Then, to control the lateral shrinkage (which is the most delicate for subsequent fabrication steps) the most important factors were found to be stack thickness and surface area. However, we already have decided to keep the surface area high and the stack thickness cannot be freely varied since it depends on the design, thus only one significant factor remains to control this x- and y-shrinkage, i.e. the temperature. Yet, the effect of this factor is limited and cannot be used to counter the impact of the stack thickness. Hence, the best way to implement these results is to go by the following steps:

- Always use the same overall surface size
- Design the device in scale 1:1 (several devices may be implanted on the overall surface)
- Decide on a fixed pressure and temperature value for your production and calculate the vertical and lateral shrinkage by (1) and (2)
- Compensate the design files with respect to the calculated lateral shrinkage value
- Fabricate the device accordingly

V. CONCLUSIONS

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 are significant factors. The vertical shrinkage can be predicted by the model (1). 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. These results are somewhat contradictory as, compared to [5] and [6], lamination pressure and temperature in the first case and lamination pressure and metal loading in the second case were found significant, out of their tested factors for the lateral shrinkage result parameter.

During this work we have established two shrinkage models, (1) and (2), that can be used to predict the shrinkage and thereby calculate the design compensation that is needed to achieve the final size after firing as intended. Surprisingly, the lamination pressure and temperature have little impact on the lateral shrinkage result, at least within the limits tested in this work and cannot really be used to fine tune the shrinkage.

This result from this work underlines the importance of each tape user to realize their own parameter setting analysis in order to adjust it to the usage and equipment at hand.

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