Influence of Lamination Parameters on LTCC Shrinkage under Unconstrained Sintering

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Abstract

LTCC (Low-Temperature Cofired Ceramic) has attracted considerable interest as a material for sensors and microfluidic circuits, for which dimensional accuracy is essential. The irregular shrinkage behaviour of LTCC tapes must be taken into account when designing devices. Furthermore the shrinkage observed in practice can be different than that given by the manufacturers. This study analyses and models with Design of Experiments the shrinkage of DuPont *GreenTape™ 951AX LTCC foils. Unlike most past studies, which concentrated on firing conditions or paste compositions, only the influence of the most obvious lamination parameters is characterised in this paper. A linear model is proposed and the relative importance of the parameters discussed. The most important (lamination pressure and temperature) play a non-negligible role (up to 1% of linear dimension) on shrinkage for a given firing profile. As expected, the more pre-densification the LTCC receives during lamination, the less it shrinks during firing.*

Key words: LTCC, unconstrained sintering, shrinkage modelling, lamination influence, Design of Experiment

Introduction

LTCC technology, originally intended for high-density, high-frequency and automotive circuits, is attracting more and more interest in other applications such as sensor technology and microfluidics. For purely electronic circuits, dimensional accuracy is mainly an issue for processing and packaging. On the other hand, sensors and microfluidic circuits that directly use LTCC require accurate control of their absolute dimensions for proper functioning. Unfortunately these new applications come up against new problems.

Firstly, as for all LTCC applications, the main issue is the shrinkage during sintering (~10-15%). However the effects of deviating from the standard parameters during lamination are not known. For instance, in production, it would be very interesting to shorten this process, but the LTCC tape manufacturers provide no information about this. Furthermore, we often encounter a different shrinkage than the values they provide is often obtained, despite carefully following their manufacturing guidelines.

Secondly, all circuits involving empty cavities face a real dilemma: the sagging of the walls (bridge effect) and delaminations between layers. As can be seen on Figure 1, channels get crushed during lamination (with a vertical uniaxial press) because there is nothing to sustain the forces acting on the walls, and the recommended temperature (70°C) softens the LTCC. Moreover, the lower stresses around cavities result in poor lamination, local variations of shrinkage and delaminations in the adjacent layers as well as in the corners of cavities.

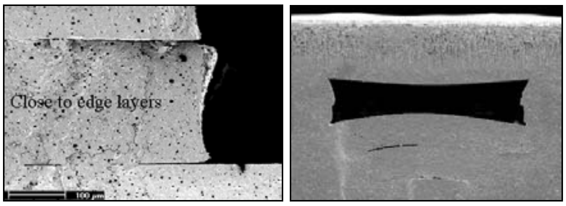

Figure 1: Left: delaminations between layers in corner of cavity. Right: SEM cross section of a 1x0.42 mm 2-layer channel showing sagging and delaminations (laminated at 70°C - 200 bars).

Thus, choosing the right parameters for processing microfluidic circuits and predicting their final dimensions with good accuracy is difficult and it is critical to take into account the impact of shrinkage variations (SV) on design considerations.

The problem of cavity integrity will be the object of future work. In this paper we study the influence on shrinkage of the pre-firing parameters, i.e. most obviously the lamination: its temperature, duration, pressure, as well as the number of LTCC layers. For this purpose experiments on test samples are conducted using Design of Experiments with a minimised number of runs, in order to directly obtain an evaluation of the importance of all parameters, their mutual interactions, as well as the quality of measurements.

We chose a linear model. Some parameters can be neglected and we propose a simplified model.

The challenges

We are using *DuPont* GreenTape™ 951AX LTCC 6"x6" 254µm-thick foils to manufacture 50x50 mm fluidic circuits with up to 22 layers and containing channels up to 3 mm wide, without co-firing screen printed films. The X-Y shrinkage is given at 12.7 ± 0.3%, and in Z at 15 ± 0.5%[1]. Upon delivery of a LTCC lot, its X-Y shrinkage is certified to 0.1%, so circuit raw dimensions must be adapted frequently. Our setup and process are described later. Our main problems are:

1) When following the manufacturers processing guidelines for lamination (70°C – 200 bars – 10 minutes), the shrinkage is different than predicted and presents up to 3% of anisotropy. We almost always observe a shrinkage greater than 13%, instead of less than 13%. As a reminder, shrinkage for standard commercial tapes[2] (under unconstrained sintering) is in the range from 9.3 to 15.3% in X-Y with variations of up to ±0.3%, and in Z (along the lamination axis) from 10.5 to 24% with variations up to ±1.5% [1].

2) The channels >1 mm wide get irremediably crushed with the same set of lamination parameters. The top and bottom layer are either touching each other or get perforated. Narrower channels are usually not subject to bridge effect, but can still suffer from delaminations.

3) Apart from using sacrificial carbon inserts, the crushing of channels suggests reducing the lamination, i.e. reducing pressure, temperature, and/or duration. This is what we have tried, with unsuccessful results. (Figure 2)[3] The problem is just shifted, with less sagging but more delamination, as well as an even greater shrinkage (up to 14%). This latter point is not surprising; our former experiments proved that less dense LTCC (i.e. less laminated) shrinks more upon firing.

The shrinkage behaviour of ceramics depends on many parameters including particle size and shape distribution, as well as size and mass of the package laminate. Most of the past studies are oriented on paste composition, thick-films compatibility, firing profiles, constrained sintering or visco-elastic modelling focussing on the molecular level [2], but few or none discuss the effects of lamination from a circuit manufacturer's point-of-view. Indeed, most experiments are run with the lamination parameters recommended by the manufacturer, or with slightly diverging "home recipes", but there are no indications about the consequences of deviating from these recommendations.

Therefore, before investigating new techniques to improve the integrity of the channels, we prefer to determine the influence of lamination parameters on X-Y shrinkage. Shrinkage in Z is less problematic because it does not cause misalignments upon further processing like screen printing.

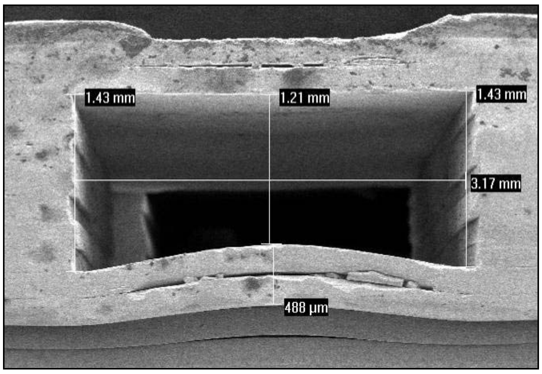

Figure 2: SEM cross section of a 3x1.47 mm 7-layer channel crossing another channel and showing sagging and delamination (laminated at 25°C - 160 bars - 10 minutes).

Parameters influencing LTCC shrinkage

The parameters that can possibly influence the LTCC shrinkage are numerous. Here is a probably non-exhaustive list, in the manufacturing chronological order:

- ageing of LTCC sheets (age, atmosphere humidity and temperature)
- method of removing Mylar backing tape (by hand, with vacuum setter, before of after blanking)
- preconditioning (before or after blanking, with temperature and duration)
- blanking method (laser or punching)
- type of release tape used for lamination (Mylar, Tedlar, new or reused sheets etc.)
- layers stacking method (alternated or not[4], numbers of layers)
- lamination technique (type of press, pressure, temperature, duration)
- elapsed times (before removal of release tape, and then before firing)
- firing method (belt of box furnace, type of setter[5], profiles for burnout and sintering (ramps and peak temperatures), position in the furnace)
- type and flow of firing gas (air, N_2, Formagas, H_2 etc.)

Design of Experiments plan

Due to the long firing profile (8 hours) and the number of parameters involved, we decide to concentrate on only four parameters: number of LTCC layers (n), lamination pressure (p), temperature (T) and duration (t). It is also decided to measure the samples dimensions at three distinct

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[1] For a stack of 8 layers, lamination 206 bars, 70°C, 10 minutes.

[2] The zero-shrinkage *Heraeus* HeraLock® HL2000 tape has not been taken into account.

[3] The sample of Figure 2 has not been polished after diamond saw cutting and the black spots are finger prints.

[4] LTCC sheets have an orientation because they are tape casted.

[5] Sunappan et al. have observed a dependence for peak temp. [3]

stages to observe two effects: 1) expansion from lasered green tape (state A) to lamination (state B), and 2) shrinkage from lamination (state B) to firing (state C). The state A is necessary because the laser presents some variability in the cutting dimensions.

The Design of Experiments plan chosen is a Linear Full Factorial Design with central point. However quadratic terms cannot be added, for the factorial design does not allow it. The central point will just allow verifying the "lack of fit". The orthogonality does not need to be verified because this plan is orthogonal by definition.

Parameters discussion

The experiment parameters will vary according to the Table 1. The reasons are as follows:

Table 1 – Experiment parameters

	T [°C]	t [min]	p [bar]	n
min	25±1	5±0.1	80±7	3
central	40±1	15±0.1	190±7	6
max	55±1	25±0.1	300±7	9
DuPont	70	10	206	(>=8)

- Temperature (T): too low (ambient) and the layers interpenetrate badly. Too high (>70°C) and the LTCC softens so much that it creeps and the channels get crushed during lamination. Note that our max T reaches 55°C only instead of the recommended 70°C. This is because we first tried at 70°C and 300 bars and our test samples were so damaged that they were barely measurable.
- Duration (t): time is potentially less important than the other parameters. *DuPont* recommends 10 minutes but we found from 5 to 15 minutes in the literature without any explanation. We decided to start the timer once the pressure reached the desired value (p hold for 2 minutes, see later).
- Pressure (p): this parameter seemed to play the most important role on shrinkage and lamination quality. Too low (<80 bars) results in bad lamination. Too high (>200-300 bars) and channels get crushed. If p is applied unequally, a trapezoidal deformation or general curvature of the samples is generated, despite no external indices after lamination. Therefore a high-quality alignment fixture is paramount.
- Number of layers (n): due to the inhomogeneity of LTCC sheets (in thickness with the grains orientations, and in X-Y with the tape casting), the number of layers could influence the shrinkage by the rubbing against the alumina substrate and rubbing between the layers themselves. Circuits with cavities require at least three layers (w/o using sacrificial carbon or organics); below that the warpage is almost inevitable anyway.

Based on previous observations and theory, the parameter dependencies are expected to be: 1) for expansion by lamination: increase with T, p, t; 2) for shrinkage by firing: decrease with increasing T, p, t. The effects are unknown for n.

LTCC test samples

Test samples of 72x10 mm have been designed to allow two distance measurements in X and two in Y per piece with Ø2.5 mm circular-through holes (Figure 3). It is done with the help of a vision system by blob detection that allows precise determination of the centre of each hole.

Figure 3: "X-ray" sketch of LTCC test samples.

In raw dimensions, $X_1 = X_2 = 50$ mm and $Y_1 = Y_2 = 5$ mm. The Y measurements are for anisotropy determination, priority is given to X. To minimise distortions, the pin alignment holes are placed at the extremities of the samples. To ensure that channels would not be crushed by the test conditions, two simple 1.5 mm-wide channels are placed between the measurement and the alignment holes; it is then easy to verify their integrity already after the lamination (Figure 4).

Two samples are prepared for each set of parameters. This is also due to our alignment fixture, which requires an even number of samples to balance the load (Figure 5). Up to 10 samples can be laminated simultaneously thanks to intermediate metal plates.

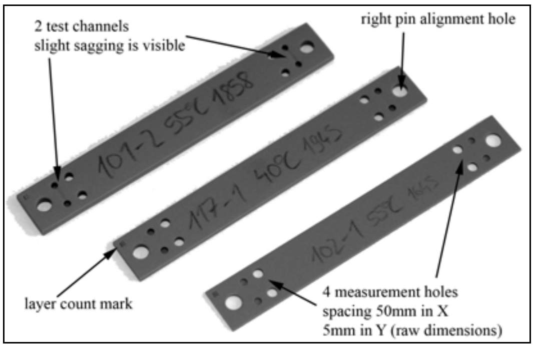

Figure 4: LTCC test samples (9, 6 and 3 layers).

Experimental setup

A package of LTCC *DuPont* 951AX containing 100 6"x6" sheets has been used for our experiments; its shrinkage was rated at 12.7%. The packet had been opened for 3 months and was kept in room atmosphere (23-25°C, 40-50% RH) in its plastic bag. The experiments occurred all during the same day (from preconditioning to firing).

Each sheet had its Mylar backing tape removed and immediately put in a drying oven at 120°C for 30 minutes for preconditioning. Then, the structuration was done by laser[6] with the sheet orientation mark pointing down. The dust generated by the cutting was removed by air blowing and soft brush. For the state A, top layers were measured with the camera of the laser system. The vision system has a measurement repeatability of $\sigma_{mes} = 3\mu m$, i.e. 0.006% over 50 mm.

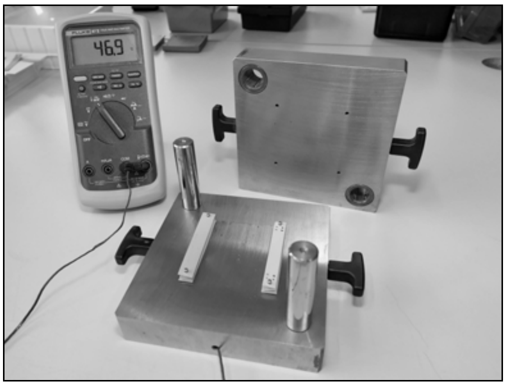

Figure 5: Test samples collated and stacked on pins of alignment fixture, just before lamination.

Our home-made alignment fixture (Figure 5) was pre-heated in a drying oven at 25, 42 or 58°C to respectively reach 25, 40 or 55°C on the LTCC, accounting for a slight cooling during manual stacking and lamination.

Collating and stacking occurred as fast as possible. The release tape used to avoid LTCC from sticking to metal walls was *DuPont* Tedlar. The same Tedlar layers were reused many times, as they did not suffer much from deformations.

Figure 6: Alignment fixture on uniaxial press.

To allow LTCC reaching the desired *T*, the whole stacked alignment fixture was placed again in the drying oven for 5 to 10 minutes. After removal of the mobile pins, it was placed in a uniaxial press on 3 mm-thick rubber discs acting as buffers against pressure imbalances and heat losses. The pressure was gradually increased until reaching the nominal value ±7 bars, then the timer was started and the pressure manually maintained for 2 minutes before releasing the press lever. The pressure then slightly decreased due to creep. (Figure 6)

After lamination the Tedlar release tape was promptly removed and the second measurements carried out. Test samples were subsequently laid on 0.6 mm-thick, 96% alumina substrates and surrounded by small alumina pieces before firing at the end of the day. The repartition of the samples on the substrates was pseudo-random (spread over floors and opposite left-right).

Table 2 – Firing (oven) profile in air at 400 l/h

Step		Duration [h:min]	Total time [h:min]	Final temp [°C]	Slope [K/min]
1	Fast ramp	00:25	00:25	230	8
2	Ramp to **440**°C	01:30	01:55	450	2.4
3	Burnout dwell 100 mins	01:39	03:34	450	0
4	Fast ramp	00:21	03:55	660	10
5	Sintering ramp to **875**°C	01:35	05:30	895	2.5
6	Sintering dwell 30 mins	00:30	06:00	895	0
7	Natural furnace cooling	00:30	06:30	400	-16.5
8	Fast cooling	00:10	06:40	200	-20
9	Back to ambiant	00:10	06:50	70	-13

The firing occurred in an IR lamp heated quartz tube furnace[7] under an air flow of 400 litres/h during all the process. The Table 2 lists the firing steps. Note the slight discrepancies between the oven and the actual LTCC temperature. We ran numerous tests with three thermocouples to verify the inside temperature, one on each substrate, in contact with a sample. We observed up to 7 K difference between the floors during the burnout dwell and only 3 K during the sintering (Figure 7).

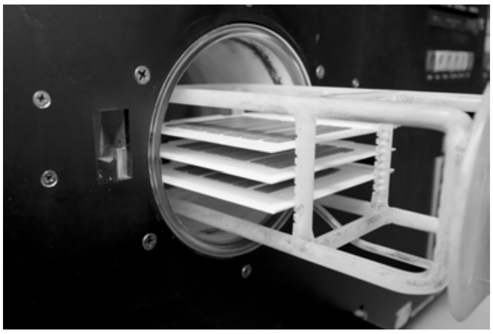

Figure 7: Fired test samples loaded on 3 alumina setters exiting the lamp furnace on the quartz substrates-carrier.

After sintering, the oven was naturally cooled until reaching 400°C, whereupon an additional air flow of 2000 l/h accelerated cooling down faster. At this stage it should play no role in the quality of the samples. The samples were then measured one last time.

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[6] LS-2000 from LS Laser Systems, Münich. Laser LS-520G, Nd:YAG with Q-switch, λ=1064μm, P=3W monomode "TEM00"

[7] PEO-601 from ATV Tech, Münich

Hypothesis

The following assumptions have been made:

- LTCC ageing plays no role on shrinkage.
- LTCC sheets were of identical properties.
- Reusing Tedlar release tapes plays no role.
- Firing profile is slow and long enough to ensure good organics burnout and sintering homogeneity for all the samples.
- Slight differences of temp. inside the oven are negligible, and firing runs are always similar.

Results – initial experiments

Initially we planed to analyse the two sub-models ABx and BCx, but the lamination process (ABx) is so variable that no relevant information can be extracted. To the contrary the BCx sub-model is nearly the same as the ACx model, so we concentrate on this latter. The measurements results have been processed using complete model matrices with interactions.

Figure 8 presents the parameters influence in regard to the constant value (which is 13.48% of shrinkage, i.e. 6.74 mm). The lamination parameters have little influence relatively to the constant. *p* is the most important one (-3.2%), followed by *T* (-1.8%), and *T*p* (-0.7%). Surprisingly *t* and *n* do not play a big role, with -0.2% each (the interaction *T-t* is even greater with 0.3%). All other interactions are smaller.

It is interesting to note that the influence of *n* in ACx only comes from ABx, because *n* has an effect of nearly zero in BCx (not displayed).

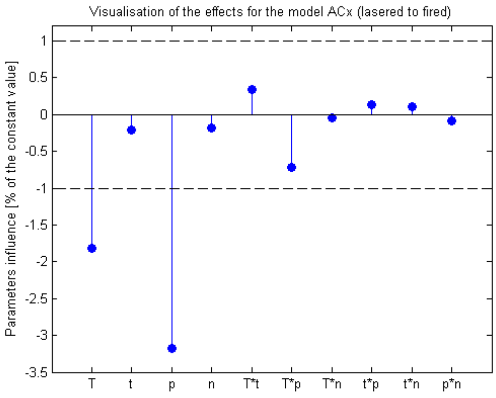

Figure 8: Influence of the different parameters and their interactions for the model ACx (from lasered state to fired state). Constant=13.48%.

These initial results confirm our expectations: more pre-densification provided during lamination lessens the shrinkage upon firing. The also suggest that simplifications in the ACx model can be done. Thus we decided to retain only the effects greater than the dashed lines (threshold at 1%) on Figure 8, *T* and *p*, and to redo the experiments but with more points.

Results – second experiments

To test the reproducibility of the initial experiment, a second run was conducted four months later with a second package of LTCC (shrinkage rated at 12.5%, package opened four months before). This time only *T* and *p* were varied, with $t = 5$ [min], $n = 3$ layers and more intermediate points: *T* [°C] = [25;40;55], *p* [bar] = [80;190;300]. In DOE this is a composite design with N=2.

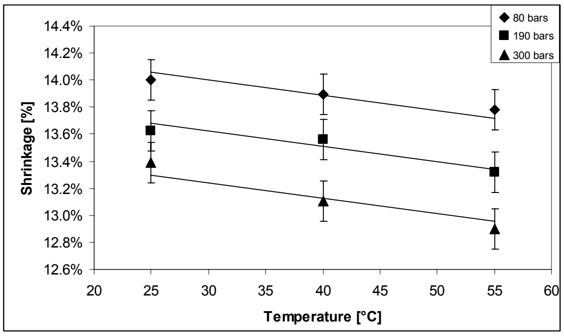

Figure 9: Experiments of ACx shrinkage (from lasered to fired state) in function of (*T*, *p*), as well as model (lines). Measurements error bars = 1 σ.

The results are presented in Figure 9. The pressure *p* has a relative influence of -2.38% of the mean shrinkage, and -1.3% for *T*. The shrinkage is presented as a function of lamination temperature for three lamination pressures. Up to three experiments were conducted for each point of the graph, and the measurements error bars displayed represent 1 σ (standard deviation) of the process variability (0.15[%]).

It is interesting to mention that the variability between two experiments of same parameters is 2 to 5 times bigger than the variability between the two samples of a single experiment (fired at the same time). This is because the former contains the operator variability, as well as the LTCC inhomogeneities if the samples belonged to different sheets of tape.

Model of shrinkage

Despite some overlapping of the error bars, it is clear that there is a distinct dependency of *T* and *p* on shrinkage. It ranges from 13% to 14%, which is not negligible. A linear regression has been performed; our model of shrinkage is displayed on Figure 9 by three lines, one for each lamination pressure. Its equation is, after de-normalisation and for *T* in [°C] and *p* in [bar]:

$$f_{ACx}[\%] = 14.62 - 1.13\cdot10^{-2}\cdot T - 3.45\cdot10^{-3}\cdot p$$

The linear model fits the points of the graph well. We have also accomplished an ANOVA analysis to assert the pertinence of our model: its Fisher P-factor[8] amounts to $3.0*10^{-12}$, and its R^2 = 0.999985. However, the R^2 is of little significance in our case, because our constant value is much bigger than the half-effects and biases the result.

The parameters have also been scrutinised individually: the P-factor for $T = 5.8*10^{-4}$, and for p it is $6.1*10^{-6}$, which is very good. We also tried a model that includes the interaction $T*p$ (as it was the third most important on Figure 8), but the P-factor of $T*p$ was 0.63, which indicates a large uncertainty on this parameter.

It is interesting to compare our model with the data from *DuPont* for our lot of LTCC (12.5% of shrinkage): with T = 70°C and p = 206 bars we find a shrinkage of 13.11% instead of 12.5% as announced. Whereas this seems to confirm that the shrinkage is greater than proclaimed, we must be careful in applying our model beyond our maximum temperature, as the binder properties are expected to become nonlinear. *DuPont* recommends t = 10 [min] and we used 5 [min], but as we have seen it plays no role on shrinkage.

Regarding the X-Y anisotropy, Figure 10 regroups all second experiments. As a general rule a linear tendency is observed, albeit with a rather high scatter in the low shrinkage area. Especially, the points for 55°C - 300 bars present more dispersion than the others. Nevertheless, a linear regression has been drawn (with intercept set at zero), of which the equation is: Y-shrinkage ≈ 95% of X-shrinkage, with a sigma of 1.55%.

However the R^2 for this model is not satisfactory and more careful experiments should be done to refine it (notably by measuring the Y-shrinkage over the same distance as in X).

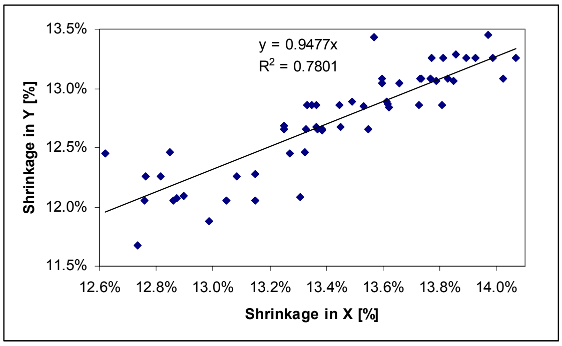

Figure 10: Measures of AC shrinkage (from lasered state to fired state) in X and Y to estimate anisotropy.

Conclusions

The goal of this study was to analyse and model with Design of Experiments the shrinkage of *DuPont* GreenTape™ 951AX LTCC foils. All the steps of our process are presented in detail. Taking into account that the usual LTCC shrinkage repeatability is around 0.2-0.3%, our process seems to be under control with a variability of 0.15 [%] (1σ).

We conclude that the number of LTCC layers (n) and the duration of lamination (t) can be neglected to predict the shrinkage. Only lamination pressure (p) and temperature (T) are retained for our linear model. Its P-factor amounts to $3.0*10^{-12}$, which proves that the model fits the reality very well. As expected, the more pre-densification the LTCC receives during lamination, the less it shrinks during firing.

For our LTCC batch, in relation of the mean shrinkage, we found that p has a relative influence of -2.38%, and -1.3% for T. This translates into a change of -0.38% and -0.17% of the absolute shrinkage values. Our model of shrinkage applied to manufacturers' conditions is larger than what *Dupont* claims (13.11% instead of 12.5%), and more coherent with values encountered in literature.

Such a model could be useful to define a standard of shrinkage prediction that manufacturers should provide with each batch delivered. It allows laminating under a broader set of conditions, instead of the one recommended by the manufacturer.

Although the shrinkage is now better understood, integrity of cavities is still unsatisfying and the process is long. Therefore we want to investigate next new methods of lamination, as well as to optimise the firing profile to obtain shorter times and a better quality.

Acknowledgments

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[8] The Fisher P-factor should ideally tend towards zero.