Non-Newtonian fluid flow model for ceramic tape casting

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Abstract

The current device of miniaturisation and higher device counts in integrated circuit (IC) packages has significantly increased the use of both multilayer ceramic packages (MLCP) and multilayer capacitors (MLC). Currently, one of the main methods used for the manufacture of flat ceramic packages with precise thickness control and consistency is the tape casting technique. Since these tapes can be cast with thickness of about 100 \(\mu\)m, it is crucial that the control of green tape thickness is precise, and that these thickness values are reproducible consistently. The flow of the slurry onto the casting surface can be modelled as a two dimensional fluid flow through a parallel channel. By choosing a suitable constitutive model, the predictions of the proposed model and existing models were compared with experimental results. The proposed model accurately described the fluid flow characteristics of the process, and had good agreement with experimental results. © 2000 Elsevier Science S.A. All rights reserved.

Keywords: Tape casting; Doctor blade; Fluid flow; Non-Newtonian

1. Introduction

The current device of miniaturisation and higher device counts in IC (integrated circuit) packages has significantly increased the use of both multilayer ceramic packages (MLCP) and multilayer capacitors (MLC). Ceramic co-fired multilayer packages form the basis on which integrated circuit packages are derived. These packages may take many different forms, such as dual-in-line packages, chip carriers, pin grid arrays and ball grid arrays. The semiconductor devices are housed in a strong, thermally stable and hermetic environment. Currently, one of the main methods used for the manufacture of flat ceramic packages with precise thickness control and consistency is the tape casting technique [1]. This method utilises a specially formulated slurry, which is cast by a doctor blade onto a flat sheet or carrier, and then dried into a flexible green tape. This green tape is then laminated into a multi-layered structure after the conductive circuits have been deposited. The whole structure is then co-fired (sintered) in a protective atmosphere of either \(N_2\) or \(H_2\) into a hard multi-layered ceramic package.

Conventional ceramic processing techniques such as dry pressing, plastic moulding, extrusion, and slip casting are not suitable for the preparation of thin, wide ceramic sheets with smooth surfaces, precise dimensional tolerances and adequate green strength for handling [2]. Thus, tape casting has become the basic process for meeting the needs of the electronics industry [3].

There are generally two types of tape casting benches: continuous and non-continuous types. Non-continuous benches are generally small benches that are used in laboratories [4]. The casting head (tank) and doctor blade traverses over a fixed support, which is often a piece of glass slab (sometimes coated with a plastic film). This is driven by either an electric motor or a pneumatic system with speed control. After drying, the tape is stripped off the glass slab. In the continuous type casting bench [1], the moving carrier is a flexible, endless belt of stainless steel or plastic films: Mylar, cellulose triacetate or polyethylene. The slurry is pumped from a slip chamber to the casting head behind the blade, keeping the feed level constant. The moving carrier is motor driven under a fixed doctor blade, and it pulls a thin layer of slurry along with it.

The applications of tape cast products are found in a wide variety of industries, such as microelectronics
Fig. 1. Schematic view of a tape casting unit.

provides a better description of the flow characteristics of tape casting. As an example, by choosing a suitable constitutive model, the predictions of the proposed model and existing models are compared with experimental results.

2. Analysis

Fig. 2 shows the relationship between the various constitutive flow behaviours. Rheological experiments were first performed on the casting slurry to determine the fluid behaviour. A Carri-Med controlled stress rheometer was used to test the slurry properties under shear. It has been shown previously [16] that the slurry did not behave according to the Newtonian or Bingham plastic models, but as a non-Newtonian pseudoplastic fluid following the Ostwald de Waele power law constitutive model. Fig. 3 shows this relationship. It is worthy to note here that the Herschel–Bulkley model is an extension of the power law model in that a yield stress, \( \tau_y \) is introduced:

\[
\text{Ostwald power law model} \Rightarrow \tau = k \left( \frac{\partial u}{\partial y} \right)^n
\]  

\( \text{(1)} \)

As can be seen from Fig. 1, the flow of the slurry onto the casting surface can be modelled as a two dimensional fluid flow through a parallel channel. A model for predicting the thickness of the green tape was first developed by Chou et al. [11] who modelled the flow of the slurry within the channel as a linear combination of a pressure and a drag flow. In their analysis, they assumed that the fluid behaved as a Newtonian fluid. However, tape casting slurries are complex multi-component suspension systems, which seldom behave as a Newtonian fluid. Furthermore, the linear summation of pressure and drag flow yields an equation where the drag flow component will increase proportionally to the casting velocity, and the effects of the pressure flow become insignificant at high velocity. Thus, the model incorrectly predicts a levelling off of the tape thickness with increasing casting velocity.

Ring [12] modelled tape casting fluid behaviour by applying the Bingham constitutive equation [13] for solving the pressure and drag flow equations. The Bingham equation represents a zone-dependent linear relationship between the shear stress and the shear rate where the slurry is assumed to flow as a Newtonian fluid in the zone where flow occurs. In his analysis, Ring made use of shear rate as a yield criterion to divide the flow and no-flow zones. A more recent model for tape casting was developed by Huang et al. [14], who utilised the Herschel–Bulkley constitutive model [15] which assumes that the fluid behaves as a viscoplastic material. Shear stress was used as a yield criterion to divide the flow and no-flow regions. However, this may not provide a good physical description, and hence not provide an accurate prediction of the flow characteristics of tape casting.

In this paper, an alternative fluid flow model for ceramic tape casting is proposed. The proposed model
where $\tau$ is the shear stress, $\partial u / \partial y$ is the shear strain rate, $k$ and $n$ are the fluid constants where $k$ represents the consistency of the fluid, and $n$ is a measure of the deviation from a Newtonian fluid. Although, the Herschel–Bulkley constitutive model is a more generalised equation, the Ostwald power law constitutive model will be employed here for simplicity. Several objections have been raised against the use of the power law model, and that other empirical equations such as the Prandtl, Eyring, Powell–Eyring and Williamson equations may be employed [17]. However, for simplicity and for the present purpose, the power law is found to be adequate. The concept, however, is not limited to the Ostwald power law equations, and can be substituted with the appropriate constitutive equations as required.

2.1. Non-Newtonian fluid flow model visualisation

Consider an imaginary tape casting system where the two tape casting chambers are a mirror image of each other, and are functioning back-to-back, as shown in Fig. 4. A pressure gradient, $\Delta P$ exists, which produces a parabolic pressure flow profile through the doctor blade region (neglecting the effects of gravity). This type of flow has been well analysed as the pressure flow of a fluid in a parallel channel [17,18].

Now, if we imagine an infinitely thin $x-z$ plane located at $y = H_A / 2$ which is moving at the maximum fluid velocity, $U_{\text{max}}$ (i.e. fluid velocity at $y = H_A / 2$). Applying a no-slip boundary condition between the fluid and the plane (i.e. both the fluid and the plane are moving at the same velocity), then the presence of the plane would not change the fluid velocity profile. The fluid would not ‘know’ that the plane is there (Fig. 5). Now, if the bottom chamber is removed, and the $x-z$ plane is replaced by a moving belt, this would represent an actual tape casting flow process where the fluid is subjected to a static pressure gradient ($\Delta P$), as well as a belt velocity of $U_{\text{max}}$ (Fig. 6).

Note that for the physical tape casting system, only half the fluid velocity profile (either the bottom or the top half) has to be considered. If we considered the top half, the volumetric flowrate would now be the integration of the velocity from $H_A / 2$ to $H_A$. Therefore, it can be seen that the fluid velocity profile of a tape casting system can be directly related to that of a pressure flow between parallel channels. A no-slip boundary condition is applied at both the fixed wall and moving belt, thus satisfying the boundary condition that the fluid next to the moving belt is travelling at the velocity of the belt.

For a Newtonian fluid, analytical solution exists. For the normal tape casting system with no-slip conditions at both the fixed wall and the moving belt [11], with a blade gap of $h_0$ and a belt velocity of $U = 1/2 \Delta P h_0^2$, the flowrate derived analytically is,

\[
Q = \frac{h_0^3}{12 \eta} \frac{\partial p}{\partial x} + \frac{U h_0}{2} \tag{3}
\]

If this imaginary $x-z$ plane is now located along an arbitrary plane within the fluid velocity profile, the plane travels at the same velocity as the fluid at the same plane so as to satisfy the requirement that there is no slip between the fluid and the plane (or the belt). This would simulate a physical casting fluid velocity profile, but with different casting parameters of pressure and belt velocity (Fig. 7). In this case, the physical doctor blade height would be the distance between the moving belt and the top channel wall ($y = H_A$). Since there is only one casting chamber, $ho$ will be used to denote the physical blade gap. $H_A$ is the gap from which the fluid velocity profile was calculated. If the
belt is now moving at slightly higher velocity, then in order to match the velocity along this plane, imaginary gap would have to be appropriately increased so as to match the fluid velocity with the velocity of the imaginary plane (the belt). A corresponding velocity profile can be derived. This profile is then integrated from the imaginary plane (the belt) to the top channel wall to obtain the physical fluid volumetric flowrate. Therefore, by maintaining a known hydrostatic pressure and a known belt velocity, the fluid velocity profile and volumetric flowrate can be accurately obtained.

Since the tape casting slurry was found to behave according to the Ostwald (Power law) constitutive equations, it has the following relationships [17]:

Applying this to the tape casting system, the fluid velocity profile is derived:

\[
\tau = k \left( \frac{\partial u}{\partial y} \right)^n \quad \text{where } n < 1
\]

(4)

\[
\mu = k \left( \frac{\partial u}{\partial y} \right)^{n+1}
\]

(5)

\[
\frac{\partial p}{\partial x} = k \left( \frac{\partial u}{\partial y} \right)^n
\]

(6)

Applying this to tape casting system, the fluid velocity profile is derived:

\[
u = \frac{y(\partial p/\partial x) - H_{\lambda}\partial p/\partial x)^{1/n+1} - \left[ -H_{\lambda}\partial p/\partial x \right]^{1/n+1}}{(\partial p/\partial x)(1/n+1)k^{1/n}L}
\]

(7)

where \( L \) is the channel length.

Now, the mass flow rate \( (Q_m) \) of the slurry is,

\[
Q_m = \rho_s Q_v
\]

(8)

where \( \rho_s \) is the slurry density and \( Q_v \) the slurry volumetric flowrate (integration of velocity, \( u \)). Now, the slurry mass flow rate is identical to the deposition rate of the ceramic tape onto the carrier,

\[
Q_m = \rho' \delta' WU
\]

(9)

where \( U \) is the casting velocity, \( \rho' \) the wet tape density and \( \delta' \) the wet tape thickness.

For actual tape casting, a side flow will take place as soon as the slurry flows onto the open surface. This extends the width \( W \rightarrow W' \) and reduces the thickness \( \delta' \rightarrow \delta'' \). Since the density remains constant at this point, we have,

\[
W\delta' = W''\delta''
\]

\[
\delta'' = z\delta'
\]

(11)

where \( W'' \) is the extended width, \( \delta'' \) the reduced thickness and \( z \) the correction factor for side flow \( (W' / W'') \).

\[
\delta'' = \frac{z\rho_s Q_v}{\rho' WU}
\]

(12)

After the slurry is cast, the solvents evaporate and the tape ‘solidifies’. Weight loss occurs at this stage and the tape thickness decreases as the tape densifies. The correction factor, \( \beta \) will be used to take this weight loss into account.

\[
\delta_p = \frac{z\rho_s Q_v}{\rho_W WU}
\]

(13)

where \( \beta \) is the correction factor for weight loss during drying, \( \delta_p \) the dry tape thickness and \( \rho_W \) the dry tape density. As observed during actual tape casting, lateral shrinkage was negligible in relation to the casting width, and will be omitted in the calculations.

The new thickness of the green dried tape is,

\[
\delta_p = \frac{z\beta \rho_s Q_v}{\rho' WU}
\]

(14)

2.2 Non-Newtonian combination of pressure and drag flow model

For comparison, the linear superposition method for pressure and drag flow was used to derive a separate model using the Ostwald power law constitutive model. This analysis was also based on the assumption that the flow is fully developed at the exit of the channel. The viscous drag component was taken as the relative velocity between the fixed doctor blade and the moving belt. The total volumetric flow rate was assumed to be a linear combination of a fully developed pressure and
drag flow, as derived from the power law constitutive model. All other system flow assumptions were kept the same. The volumetric flow rate as contributed by the pressure flow is denoted by $Q_p$, volumetric flow rate as contributed by the drag flow is denoted by $Q_d$, and total volumetric flowrate is denoted by $Q_t$.

Using a similar approach by other investigators for pressure and drag flow [11], it can be shown that,

$$Q_t = Q_p + Q_d$$

$$= -\frac{2Wp}{L} \left( -h_0/2\right) h^1/n + 2\left(\hat{c}p/\hat{cx}\right)^1/n + 1 \left(\hat{c}p/\hat{cx}\right)^1/n + 1 + \frac{1}{2} \left(WUh_0\right)$$

The thickness of the dried green tape is,

$$\delta_p = \left[\frac{2(h_0/2)^1/n + 2(\Delta p)^1/n}{L(1/n + 2)k^{1/n}U^1/n} + \frac{1}{2}(h_0)\right] \rho_s x \beta$$

3. Experimental procedure

Tape casting was performed using the formulation as listed in Table 1 below. A dual solvent system was used to improve organic solubility, and prevent preferential volatilisation and polymeric surface skin formation.

<table>
<thead>
<tr>
<th>Component</th>
<th>Function</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminium oxide</td>
<td>Ceramic substrate</td>
</tr>
<tr>
<td>Magnesium oxide</td>
<td>Sintering additive</td>
</tr>
<tr>
<td>Trichloroethylene</td>
<td>Solvent</td>
</tr>
<tr>
<td>Ethanol</td>
<td>Solvent</td>
</tr>
<tr>
<td>Corn oil</td>
<td>Deflocculant</td>
</tr>
<tr>
<td>Polyethylene glycol</td>
<td>Plasticiser</td>
</tr>
<tr>
<td>Benzyl butyl phthalate</td>
<td>Plasticiser</td>
</tr>
<tr>
<td>Polyvinyl butyral</td>
<td>Binder</td>
</tr>
</tbody>
</table>

A full description of the tape casting procedure can be found in [19].

Tape casting was performed on a batch process type caster, where the casting head and doctor blade traversed over a stationary floating glass slab, discharging slurry onto the surface (Fig. 8). The doctor blade gap was varied by a micrometer screw gauge, which could achieve an accuracy of up to 0.01 mm. Accurate casting velocity control was achieved via a stepping motor controlled by a motor controller.

Tapes were cast using constant blade gaps with varying casting speeds. The slurry formulation and all other processing parameters such as casting temperature and pressure for the casts were kept constant. Accurate tape thickness measurement was achieved by sandwiching the tape between two pieces of thin glass of known thickness, and using a micrometer to measure the total thickness. This method of measurement eased pressure from the micrometer jaws, which might cause that small portion of tape in contact with the micrometer jaws to compress and give inaccurate results.

4. Results and discussions

From rheology experiments [16], the tape casting slurry was found to behave according to the Ostwald power law fluid. The relationships obtained were:

$$\tau = k(\hat{c}u/\hat{cy})^n \quad \text{where} \quad n < 1 = 1.0357(\hat{c}u/\hat{cy})^{0.6748}$$

$$\mu = k(\hat{c}u/\hat{cy})^{n-1} = 1.0357(\hat{c}u/\hat{cy})^{-0.3252}$$

where $n = 0.6748$ and $k = 1.0357$.

For power-law fluid behaviour, the constant ‘$k$’ represents the consistency of the fluid, and ‘$n$’ is a measure of how much the fluid deviates from a Newtonian fluid. A more detailed explanation on non-Newtonian fluids can be found in [20].

Values of the various physical parameters were substituted into the relevant equations and a relationship between blade gap and casting velocity was obtained. The constant for side flow ($x$) was obtained by a volumetric comparison of tape which flowed outside the casting width to tape within the casting width. The drying weight loss factor ($\beta$) was calculated from drying experiments which measured dried tape volume and compared it to wet slurry volume.

The experimental results were plotted together with:

1. Newtonian (linear superposition) model (with $n = 1$) [11],
2. non-Newtonian (power law) model,
3. a separately derived power law model based on linear superposition.

Fig. 9 shows the results of a 0.25 mm blade gap setting, and Fig. 10 shows the results of a 0.4 mm blade
gap setting. As shown in the plots, the linear superposition Newtonian model predicts a tape thickness that levels off with increasing casting velocity. From the equations, the drag flow component will increase proportionally to the casting velocity, and the effects of the pressure flow will thus be less significant at high velocity.

From observations of tape casts, this was not true as the volumetric flowrate was observed to be dependent on the pressure supplied by the reservoir. If a fixed pressure is maintained, there will be a limiting velocity where the deposition rate of the slurry would be less than the casting velocity, and this resulted in a decreasing tape thickness.

The non-Newtonian linear superposition model seems to forecast a much lower thickness than the experimental values. This model also predicts a levelling off of the tape thickness with increasing casting velocity. This is also due to the assumption of linear superposition such that the drag flow component becomes the dominant component at high velocity.

The non-Newtonian power law model correctly predicts the trend in tape thickness. This model also takes into account the decrease in tape thickness at increasing casting velocities, where the increasing difference in casting velocity and slurry deposition rate become significant, and where the increasing shear decreases the fluid viscosity.

All the three models predict a tape thickness greater than the blade gap setting at a low casting velocity. This is not realistic, as the maximum achievable tape thickness in reality would have to be the blade gap setting. This arises because of the assumption that the flow rate is the same as the deposition rate even at zero velocity. However, this is not a significant limitation for practical situations.

5. Conclusions

A model for predicting the thickness of tape cast substrates using non-Newtonian slurries has been devel-
oped using the Ostwald de Waele (power-law) constitutive model. The proposed model has excellent agreement with experimental results and it is far more superior to the other models based on the assumption of linear combination of pressure and drag flow. It predicts the continuing decrease in tape thickness with increasing belt velocity, as confirmed by experimental observations. This is in contrast with the prediction of the linear superposition models that the tape thickness approaches an asymptotic value with increasing casting velocity.

References