Paper 16

Drying of a Multilayer Ceramic Shell Body

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Abstract

In the present work, a two-dimensional numerical model of heat and mass transfer for the convective drying process of multilayer ceramic mould system was developed. The governing system of fully coupled non-linear partial differential equations describing the process was derived from a mechanistic approach. A formulation including hygrothermal and moisture transport in soil was adopted as the basis for further development in this work. For validation, the calculation results for drying of a ceramic material (brick) showed that the model presented is in a good agreement with other studies that have been reported previously in the drying porous material. Further application is illustrated through the drying of a ceramic shell layer under conditions that are relevant to shell manufacture in the investment casting process.

Keywords: convective drying, multilayer ceramic mould system, heat and mass transfer, hygrothermal, moisture transport, mechanistic approach.

1 Introduction

Production of investment casting shell moulds require more than one ceramic layer to be applied onto the wax pattern to form the shell mould body. The drying process which is need as each ceramic layer is built up, is one of the critical and important stages in producing a good quality ceramic shell mould\(^1\). In fact, layer drying has a direct impact on processing time and a number of layers are added in order to get sufficient strength\(^2, 3\). If the shell is re-dipped before being sufficiently dry, its strength is reduced and there is the possibility of cracking in multilayer assemblies\(^4\). Therefore, the duration of layer drying and the final drying rate (when a seal coat has been applied) play an important role in ensuring the optimum shell strength \(^2\).
In drying a multilayer shell body, theoretically the thermal, moisture and pressure gradient are dynamically created within the multilayer envelopes[5, 6]. Thus, the correct evaluation of these thermophysical variables is essential for accurately predicting the heat and moisture transport in order to establish the optimum drying conditions. Recently, computer aided numerical method such as the finite element method (FEM), the finite difference method (FDM) and the boundary element method (BEM) lead to solutions in predicting the porous material response. Most of the research on shell making that has been carried out previously has used an experimental approach. This includes the complication of measuring moisture content, temperature and drying rate that generally respond in a nonlinear manner. Furthermore most ceramic porous materials are very sensitive to any thermophysical change which leads to sudden changes[7] that are difficult to capture accurately in an experimental setting.

Multilayer shell systems are built with similar materials. Thus it may be assumed that there is no discontinuity between each layer and there is no interface to generate a contact resistance. This is due to the high absorption of water at the dipping part of the layering cycle leading to good wetting as layers are built up. This has been demonstrated by extensive works [3, 8] on assessing fluid uptake as the dry shell is immersed in slurry within the fabrication cycle. Eliminating consideration of this interface resistance simplifies the mathematical formulation and simulation of this layering influence [9].

With the above assumption and consideration, the most widely used coupled heat and mass transfer formulation in a porous material under a temperature gradient including gas transport was solved numerically in an iterative way, using the FEM. By using a mechanistic concept along with Darcy’s law in expressing the moisture flux and the Fick’s diffusion law, this enables simulation to capture variables such as saturation (moisture content), temperature and water pressure.

2 Theoretical Formulation

The theoretical formulation of the heat and mass balance equations used here is based on the work by Ben Nasrallah[10], Whitaker[11] and others[12-14]. Based on the above analysis, the governing equations of the model are expressed in terms of the chosen state variables; pore water pressure, $P_l$, gas pressure, $P_g$, and temperature, $T$. The time derivative of moisture (liquid and vapour) transport as given below shows the gradient in liquid flux, the influence of a vapour pressure gradient and vapour bulk flow.

For the liquid: $$\frac{\partial \phi P_l v_l}{\partial t} = -\frac{\partial P_l v_l}{\partial x} - \dot{m}$$ (1)

For the vapour: $$\frac{\partial \phi P_g S_g}{\partial t} = -\frac{\partial P_g v_v}{\partial x} + \dot{m}$$ (2)
The mass balance of the liquid water and of the vapour, summed together to eliminate the source term related to the phase changes (evaporation and condensation), form the mass balance equation of the liquid phase

\[
\frac{\partial (\phi \rho_v S_v)}{\partial t} + \frac{\partial (\phi \rho_v S_g)}{\partial t} = -\nabla (\rho_v v_v) - \nabla \left( \rho_v v_v \right)
\]  

(3)

where;

\[
\rho_v v_v = \rho_v v_g - \rho_v v_v
\]

Water and gas velocity can be easily derived from Darcy’s law

\[
v_l = \frac{-K_l \nabla P_l + \nabla Z}{\mu_l}
\]

(4)

\[
v_g = \frac{-K_g \nabla P_g}{\mu_g}
\]

(5)

where \( \rho_v, \rho_g, \rho_l, \phi, S_l, S_g, m, Z \) are vapour density, gas density, liquid density, porosity, liquid saturation, gas saturation, evaporation and condensation term and the vertical elevation from a datum. Transport in the vapour state by diffusion is given by;

\[
\rho_v v_v = \frac{-\phi S_g D_{atm} v \alpha}{\rho_v} \nabla \rho_v
\]

(6)

where \( D_{atm} \) is the molecular diffusivity of water vapour through dry air and is given in reference[15]. \( \alpha \) is a tortuosity factor, \( v \) is a mass flow factor, given as in reference[16]. The contribution of vapour transport by diffusive flow as dealt with in this study, was derived by Philip and de Vries[12] which included the microscopic pore temperature gradient ([\( \nabla T \)]_a/\( \nabla T \)) by introducing the flow area factor (\( f_c \)). This derivation has been re-evaluated and explained fully by Ewen and Thomas[17]. However, Kanno et al[18] could not obtain a good agreement between predicted and experimental result from tests on clay samples using the above[17] derivation. In the latter work, they proved that the vapour flow area factor for clay material depends linearly on volumetric air content, where it gave the best result and therefore it was also implemented in this work. Simplification of the influence vapour flux (refer to the Equation (6)) into the diffusivity term having regarding to the measured parameters as derived in [12, 17, 19]gives;

\[
v_v = \nabla (D_v \nabla P_v) + \nabla \left( D_{v_l} \nabla \left( \frac{VT}{\nabla T}, f_c \right) \right) + \nabla (D_{v_g} \nabla P_g)
\]

(7)

where \( D_{v_l}, D_{v_g} \) and \( D_{v_l} \) are the diffusive term that included the vapour dispersive terms. The energy equation for the whole medium is given below;
\[
\frac{\partial}{\partial t}\left((1 - \phi)C_p \rho_c + \phi S_p \rho_c L\right) = \nabla (T \rho_c \nabla) - \nabla (\rho_c v_c + \rho_v v_v)L - \nabla \left( \sum_{i=1}^{\phi} (T - T_i) \rho_i C_{pi} v_i \right)
\]

where \( \lambda, L, C_{pi} \) are the thermal conductivity, latent heat and heat capacity of the related medium. By applying a mass balance to the flow of gaseous (dry air and vapour) mixture within the pores of the ceramic body dictates that the time derivative of the gaseous mixture content is equal to the spatial derivative of the gaseous mixture.

\[
\frac{\partial \rho_g}{\partial t} = -\nabla \left( \rho_g v_g \right) + m
\]

The pressure and density of the gas accounts for an air and vapour mixture that is dependent on the surrounding humidity and also the convection at the surface of the body. In this problem, the air portion is assumed to follow Dalton’s Law as an ideal gas.

### 3 Thermodynamic Relationship

The existence of a local equilibrium at any point within the porous system is assumed. The equilibrium vapour pressure can be approximated by Kelvin’s equation. This equation shows that the equilibrium of water vapour, \( P_v \), differs from the saturation pressure \( P_{vs} \), due to the curvature of the interface between capillary, \( P_c \) or hygroscopic water and the gas phase inside the pores of the medium. Given that

\[
(\text{rh}) = \exp\left(-\frac{P_v}{\rho_v R T}\right) = \exp\left(-\frac{P_v - P_t}{\rho_v R T}\right)
\]

where \( R_v \) is a specific gas constant for water vapour, the vapour partial pressure can be calculated as a function of local temperature and relative humidity using the relationship

\[
P_v = \text{rh}(P_{vs})
\]

where the saturation vapour pressure, \( P_{vs} \) may be estimated using the saturated vapour density as a function of temperature and is given in[20]. This has been fitted to the vapour-pressure–temperature data from the steam tables. The degree of saturation \( S_l \) (considering together a hygroscopic region and capillary water, if the latter is contained in the pores) is an experimentally determined function of capillary pressure and temperature[21]. In many hygrothermal and soil transport problems, this is expressed as [22]. These show hysteresis of the saturation curve in drying and wetting of clay soil and brick materials.
\[ S_l = \frac{(S_l)_{\text{sat}} - (S_l)_i}{(S_l)_{\text{sat}} - (S_l)_i} = \left( \frac{1}{1 + (\phi(P_c)T)^n} \right)^m \]  

(12)

In eq(12), the parameters \( \phi, n \) and \( m \) are three different porous material properties that are directly dependent on the shape of the water retention curve. \( (S_l)_{\text{sat}}, (S_l)_i \) are the saturated saturation and the irreducible saturation. The appropriateness of this will be addressed later within this research programme. The relative permeability expression for water(\( k_l \)) and gas(\( k_g \)) used Mualem’s model that has been developed for soils and implemented in hygrothermal research [23]. This was also implemented in this model. In the latter work, these relative permeability properties on the ceramic body were measured in the isothermal condition which is the condition for drying of a ceramic shell body.

\[ k_l(sI) = \sqrt{sI \left(1 - (1 - sI^{1/m})^m\right)^2} \]  

(13)

\[ k_g(sI) = \sqrt{1 - sI \left(1 - sI^{1/m}\right)^m} \]  

(14)

where \( m \) and \( n \) terms and values in the Equation (12) to the Equation (14) are referring to the same terms and values as given in the reference[23].

### 4 Material Data for the Ceramic(shell) Body

In this work the ceramic shell body was considered to have the same properties as a nonhygroscopic brick. This is because the drying of investment shell ceramic is far below the temperature at which bound water becomes a consideration [24].

<table>
<thead>
<tr>
<th>( \rho_s ) (kg/m(^3))</th>
<th>( \phi )</th>
<th>( K_s ) (m(^2))</th>
<th>( C_p ) (J/(mol K))</th>
<th>( \lambda_s ) (W/mK)</th>
<th>( L ) (J.kg)</th>
<th>( S_l )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2000</td>
<td>0.35</td>
<td>( 1 \times 10^{-14} )</td>
<td>925</td>
<td>1.8</td>
<td>( 2.4 \times 10^6 )</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Table 1: Physical and transport properties of ceramic body

where \( \rho_s \) and \( K_s \) are solid density and intrinsic permeability of solid phase

### 5 Boundary Condition

The boundary conditions reflect insulation and impermeability at all symmetry planes and convection at all exposed surfaces. Convection is captured by means of heat transfer coefficients for thermal exchange and mass transfer coefficients for moisture transport. Representative values for these parameters are readily available in the open literature [25] and these are defined for the respective case studies.
\[ J_m = h_m (P_v^r - P_s^r) \]
\[ J_h = h_f (T_v - T_s) \]

6 Solution of Governing Equations and Numerical Method

The equations have been solved as a coupled set. Each equation can be written in the general form:

\[
[C(\Phi)] \frac{\partial \Phi}{\partial t} = \nabla \left[ \sum_{i,j=1}^{3} \left( K_{ij} \nabla \Phi \right) \right] + \rho \left( \Omega \right) \nabla Z
\]  

(17)

Through application of a Galerkin weighted residual approach, this leads to a matrix representation:

\[
K(\Phi)\Phi + C(\Phi)\dot{\Phi} + J(\Phi) = \{0\}
\]  

(18)

Where for a set coupling moisture, heat and gas transport:

\[
K = \begin{bmatrix}
K_{11} & K_{12} & K_{13} \\
K_{21} & K_{22} & K_{23} \\
K_{31} & K_{32} & K_{33}
\end{bmatrix}
C = \begin{bmatrix}
C_{11} & C_{12} & C_{13} \\
C_{21} & C_{22} & C_{23} \\
C_{31} & C_{32} & C_{33}
\end{bmatrix}
\]

\[
\Phi = \begin{bmatrix}
\frac{\partial P_v}{\partial t} \\
\frac{\partial T}{\partial t} \\
\frac{\partial P_r}{\partial t}
\end{bmatrix}, \quad \dot{\Phi} = \begin{bmatrix}
P_v \\
T \\
P_r
\end{bmatrix}, \quad J = \begin{bmatrix}
J_1 \\
J_2 \\
J_3
\end{bmatrix}
\]

In which typical elements of the matrix are:

\[
C_{ij} = \sum_{s=1}^{n} \int_{\Omega^e} C_{ij} N_s N_s d\Omega^e \\
K_{ij} = \sum_{s=1}^{n} \int_{\Omega^e} K_{ij} \nabla N_s \nabla N_s d\Omega^e
\]

\[
J_i = \int_{\Gamma^e} K_{ij} \nabla N_s \nabla z d\Omega^e - \int_{\Gamma^e} N_j n_i d\Gamma^e
\]

(i, j=1,2,3) and \( \mathbf{n} \) is the outward normal vector to the element boundary, \( \Gamma^e \) of the element body, \( \Omega^e \). The equation set was solved using a coupled formulation. The system is highly nonlinear due to the impact of thermophysical properties on the matrix coefficients. This led to the application of an iterative solution strategy within each time step.
7 Result and Discussion

7.1 Validation Case Study

Two examples are solved to validate and demonstrate robustness of the code by illustrating hygro-thermal phenomena and their evolution during the drying of a ceramic shell body. The first example deals with a one dimensional ceramic brick heated from the top side, the short sides and bottom were treated as being insulated and impermeable and the heat and moisture transfer takes place at the top exposed surface only. The relevant material properties are presented in the Table 1 and the matrix is assumed to be saturated at the commencement of the drying process. Experimental data from [15] and simulation results for brick drying at 75°C with heat and mass transfer $h_T = 6 \text{ W/m}^2/\text{K}$ and $h_m = 0.008 \text{ m}^{-1}$ are shown in Figure 1 in which moisture and temperature at the mid height of the brick are depicted over time. The reference temperature and relative humidity used are 75°C and 50-60%, and the geometry was described by a brick comprising side 10.7cm long and 3.2cm thick with the above boundary condition. This enables validation against the work by Stanish et al[15].

![Figure 1: liquid saturation and temperature changing over the drying time](image)

The result shows that the moisture drops quickly at the beginning, corresponding to the constant drying rate period. During this stage the movement of liquid is maintained by the capillarity of the porous matrix. At the same time the temperature will increased gradually starting from the top surface. At this point, the internal moisture transfer to the surface and the evaporation at the surface are in equilibrium, and the free water on the surface will be evaporated steadily and continuously. Over the drying duration, the saturation level will decrease and at nearly 60 minutes, it displays the characteristics of a falling rate period where there is a reduction in the rate of moisture loss. Theoretically, the saturation level will recede continuously into
the interior of the materials and the dry zone will extend gradually. Normally this corresponds to a critical saturation of about 0.3[19] for most porous material. This shows the starting of the falling rate period. During this period the drying process will slow down and drying is now controlled by the water vapour movement. The comparison with experimental data displayed in Figure 1 is also very good, confirming the basis and quality of the simulation model that has been developed and applied in this case study. Figure 2, depicts the change in saturation over the brick height at discrete times, showing that the moisture content decreased slowly after the falling rate period.

The same pattern for temperature is also exhibited in Figure 3, showing a small increment during the constant rate period, a big increment after the falling rate period and finally stabilizing towards the ambient condition of 75°C. This is also reflected in the permeability properties in Figure 4, where the permeability change starts to show a small reduction when nearing hygroscopic saturation. Also, as can be seen from this figure, the permeability properties drop close to zero at the saturation value 0.1. This is due to failure of liquid transport by capillary action.

Figure 2: Saturation variation along the depth.
This is no longer possible below the irreducible saturation level [26, 27]. It has been found that the experimental detection of very low permeability is extremely difficult and it is generally taken as equal to zero in the drying process. Figure 4, also includes the variation of relative humidity within the porous matrix as a function of saturation. Above the critical saturation, the humidity remains close to a saturated humidity condition and below this value it showed the falling rate condition where the vapour transport mechanism plays an important role in changing the local humidity. When near to the irreducible saturation, the relative humidity just shows a very small change, indicating the minimum water content to which a material can theoretically be dried under the non hygroscopic condition.

Figure 3: Temperature variation along the depth
Figure 4: Liquid permeability and relative humidity against saturation.

Figure 5: Thermal vapour (with and without microscopic pore temperature (gradta) gradient) diffusivity against saturation

By including a microscopic pore temperature gradient and a vapour flow factor in the Equation (6), these term change the thermal vapour diffusivity coefficient ($D_{vT}$) in the vapour transport equation as shown in Equation (7). This effect can be seen in Figure 5, where the plot without the pore temperature gradient (gradta) and and flow factor ($f_c$) showed a small change in the value $D_{vT}$. The same trend can be found in [12] and also other related papers [17] that used this derivation showing an agreement with this formulation. The consequent difference is anticipated in the falling rate period where the vapour transport mechanism plays an important role in transferring moisture to the air. However, due to the small influence on the coefficient value, this effect just showed a small difference in temperature and also a small reduction in moisture content (especially after falling rate period). The small difference was not perceptible in comparison with the large scale of temperature and moisture variation. Typically the moisture difference was 0.01-0.05%, but even this difference is significant with respect to ceramic shell drying[17, 19, 28].
7.2 Two Dimensional Case Study – Single Layer Problem

The two dimensional case study represents a corner section. This is shown in Figure 6 that also summarises the boundary condition prescription. The ceramic comprises a single layer. The material properties that were used are listed in Table 1. Initially the layer shell has a saturation equal to 95-96% and a body temperature of 19°C. The saturation and relative humidity are almost constant across the body. The ambient condition applied comprise a temperature of 23°C and a relative humidity of 50%. This approximates nearly an isothermal drying process in which moisture is removed through control of ambient humidity. Examples of the two dimensional results at different times are presented in Figures 7 and Figure 8.

Figure 6: Schematic of convective boundary conditions in single first layer

Figure 7: (a) Pore water pressure(Pw) (b) Saturation(Sl)
Figure 7: (c) Permeability ($kw$) at the beginning of drying times

Figure 8: (a) Pore water pressure ($Pw$) (b) Temperature ($T$) (c) Saturation ($Sl$) (d) Permeability ($kw$)
The results of measured parameters in the first layer within the first 2 hours of drying are shown in Figure 8. As expected, the greatest changes are observed in the corner zone where the temperature reaches its maximum value, leading to the lowest saturation, water permeability and relative humidity. Saturation, permeability and relative humidity contours show very close or similar patterns due to their strong dependence on the moisture content. At the time increments, all parameters show only a small variation across the body due to the geometry of the ceramic shell layer (1mm thick) and its ability to promote moisture transport. To some extent, this reflects industrial practice where a single layer is typically exposed to a drying duration of 2 hours.

7.3 Two Dimensional Case Study – Multilayer Problem

In this case, a multilayer shell body has been defined by having a number of zones. These are shown in Figure 9 and Figure 10(c), where the most outer layer (layer (A)) is freshly dipped, followed by the others through which the saturation level falls, leading to dry zones in layers D, E and F. The saturation values given in the Figure 9 are based on the calculation from soak back studies as described in [3,8]. Thus in constructing this layered problem, Figure 9 shows an appropriate range of saturation values. The same considerations apply to the related parameter which are thermodynamically linked to the saturation level.
Figure 9: Schematic of convective boundary conditions in multilayer shell body

Figure 10: (a) Pore water pressure (Pw) (b) Temperature (T) (c) Saturation (Sl) (d) Permeability (kw)
Results at the early stage of drying (1.3 minutes and 5.5 minutes) showed the saturation is higher at the tip of corner zone, due to the fact that the thinner layers dry faster. The same evolution is followed by the interrelated parameters and this information is shown in Figures 11 and Figure 12. Temperature shows the fastest change at the outer surface of the body at beginning of drying achieving the highest value at the saturated corner. This is probably due to the higher water content at the outer zone raising the thermal conductivity within the network. In this simulation, the inner surface (first layer) is assumed to be at a slightly higher temperature compared with the outer surface at the start of drying. Later, the highest temperature is shown at the tip of the corner zone due to heat input from the two sides.
Figure 11: (c) Saturation ($Sl$) (d) Permeability ($kw$) (e) Relative humidity ($rh$) at 1.3 minutes.
Figure 12: (a) Pore water pressure (Pw) (b) Temperature (T) (c) Saturation (Sl) (d) Permeability (kw) (e) Relative humidity (rh) at 5.5 minutes.
Figure 13: (a) Pore water pressure (Pw) (b) Temperature (T) (c) Saturation (Sl) (d) Permeability (kw) (e) Relative humidity (rh) at 6.5 minutes.
As drying proceeds, results show that the moisture content moves slowly from the wet zone to the inner shell due to convective transfer at the outer surface. The same evolution of others interrelated parameters is also exhibited in Figure 13. Now the excess moisture tends to move towards the inner dryer region driven by the temperature gradient at the highest temperature. Over the drying times, as expected, the greatest changes are observed in the corner zone where the temperature reaches its maximum value, leading to the lowest saturation, water permeability and relative humidity. The final state is depicted in Figure 14 in which thermal and saturation levels are nearly uniform throughout the matrix.

Figure 14: (a) Pore water pressure (Pw) (b) Temperature (T) (c) Saturation (Sl) (d) Permeability (kw)
8 Conclusion

The aim of this study was to establish the basis of a simulation approach to represent the heat and mass transfer that occurs during the drying of layers in a ceramic shell mould. The model used is very comprehensive and accounts for moisture transport by capillary, vapour and gas transport mechanisms. Energy transfer accounts for latent heat and diffusion including the influence of local moisture content. Verification of the model has been performed on a one dimensional domain, focusing on brick drying. Through this benchmarking, the results show that the prediction of measured variables and transport properties are in good agreement with other related work. A two dimensional study is presented for conditions that are pertinent to shell drying conditions. The predicted results for single layer shell mould system show that after 2 hours drying times the shell achieves a nearly dried nonhygroscopic body. For a multilayer shell, drying to a similar state requires more than 8 hours drying time. These results also are compared well with shell drying experimental work in which later drying effects are explored.

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