A MODEL OF SLIP CASTING

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Received 27. 5. 1991

A mathematical model, considering liquid transfer as diffusion in a binary mixture, was worked out for the purpose of describing quantitatively the casting of ceramic mixes (slips) into ceramic moulds. Relationships for the calculation of the kinetics of body formation and solidification were obtained by introduction of initial and boundary conditions. In deriving the body formation and solidification model, a number of input conditions and simplifying assumptions had to be introduced. An experimental verification of an aqueous suspension of $\alpha - Al_2O_3$ showed that the model was capable of predicting the course of body formation and solidification during slip casting into a porous mould with an error of less than 9%. In this way, the use of the simplifying assumptions introduced in deriving the model has been justified.

INTRODUCTION

The course of the slip-casting process (pouring of suspensions into porous moulds) can be optimized and controlled when one knows (i) the course of the kinetics of body formation and (ii) that of the kinetics of solidification. The two items of information can be gained from the time development of concentration fields in the system which in turn can be determined by mathematical modelling of the operation. As the slip-casting technology allows bodies of arbitrary shapes to be prepared, that is bodies of simple shapes of a plate, cylinder, sphere, as well as various combinations of the basic shapes, the model must allow the required information to be also available in relation to the body shape.

Preparation of a mathematical model of body formation and solidification in the course of forming ceramic bodies by pouring suspensions into porous moulds is the subject dealt with in the present paper. The model is based on the balance and constitutive equation following from the general diffusion theory [1]. The present study was also aimed at experimental verification of the results obtained from the model, and at assessing the suitability of the simplifying assumptions introduced in the resolving of the problem.

Mathematical model of body formation and solidification

If considering only bodies of simple shapes and on the assumption that both the suspension and the body can be regarded as binary mixtures of incompressible components, then the basic balance of weight expressed in various coordinate systems can be written in the form [2,3]

$$\delta_t C = x^{-m}\delta_x(D_{ef}x^m\delta_x C) \qquad (1)$$

where x is the coordinate in the direction of the liquid transfer, t is time, D_{ef} is the effective diffusion coefficient, and m is the coefficient given by the shape of the body, i.e., for a plate, $m = 0$, for a cylinder, $m = 1$, and for a sphere, $m = 3$. If D_{ef} is independent of concentration, equation (1) acquires the form

$$\delta_t C = D_{ef}x^{-m}\delta_x(x^m\delta_x C). \qquad (2)$$

By resolving equation (2) for the selected initial and boundary conditions describing the body formation and solidification, one obtaines the time dependence of concentration profiles in the body. Equation (1) can be resolved by the numerical grid method using Crank-Nicolson's differential pattern with spatial step h and time step k in the form

$$-C_{i-1,j+1}\Omega x_i^m + C_{i,j+1}(1+\Omega x_{i+1}^m+\Omega_i^m) -$$
$$- C_{i+1,j+1}\Omega x_{i+1}^m =$$
$$C_{i-1,j}\Omega x_i^m + C_{i,j}(1-\Omega x_{i+1}^m-\Omega_i^m) +$$
$$+ C_{i+1,j}\Omega x_{i+1}^m$$

where

$$\Omega = D_{ef}\,k\,(2\,x_i^m h^2)^{-1} \qquad (3)$$

and $C_{1,j}$ represent concentrations at the nodal points of the grid.

THE INITIAL AND BOUNDARY CONDITIONS

1. Body formation

To arrive at the initial and boundary conditions, let us use the concept of body formation shown schematically in Fig. 1. In the beginning, the suspension is in contact with the porous mould in the absence of a body. A correctly prepared suspension has its liquid phase homogeneously distributed throughout its volume, so that at time $t = 0$, the state is described by the initial condition $C = C_0$. At time $t > 0$, the body begins to form as a result of the liquid being

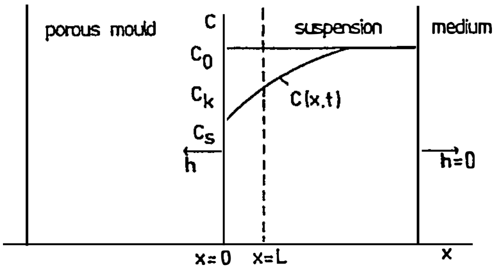

Fig. 1. Schematic representation of body formation

transported through the body-porous mould boundary. This process is described by the boundary condition $h = -D_{\text{ef}}\, \delta_x C$. In the case of pouring onto already formed body, the system contains excess suspension. If the transport of liquid phase through the suspension-environment boundary does not involve e.g. evaporation of the liquid phase, then the constancy of concentration at this boundary is described by the boundary condition $\delta_x C = 0$. The given concept may be formulated by means of the following initial and boundary conditions :

$$t = 0 \qquad x \in (0;H) \qquad c = c_0$$

$$t \in 0 \qquad x = 0 \qquad h = -D\, \delta_x C \tag{4}$$

$$x = H \qquad \delta_x C = 0,$$

where H is the height of the suspension column (head). On converting the conditions (4) into differential expression, one obtains

$$j = 0 \qquad i \in (0;N) \qquad C_{i,j} = C_0$$

$$j = j+1 \quad i = 0 \quad C_{i-1,j+1} = C_{i,j+1} + \boldsymbol{h}\, h\, D^{-1} \tag{5}$$

$$i = 0 \qquad C_{N,j+1} = C_{N-1,j+1}$$

The system of linear equations (3) obtained, following introduction of (5), is used to calculate the time development of the concentration profiles in the body. Knowledge of these profiles allows the position of the coagulation concentration at various times to be established. This position corresponds to the body-suspension boundary, and thus describes the kinetics of body formation, i.e. $L = L(t)$, where L is at the same time the position of the coagulation point and the body thickness.

Knowledge of the time development of the concentration profile, and that of the body thickness, allows the mean concentration in terms of time to be calculated according to the equation

$$\bar{C} = L^{-1} \int_0^L C(x)\, dx. \tag{6}$$

2. Body solidification

As soon as the body of required thickness has formed (i.e. after pouring the excess suspension out of the mould or after the entire volume of the suspension in the mould has converted into body), the body begins to solidify. At the onset of solidification, the distribution of liquid concentration in the body is given by its final distribution following body formation. The liquid transfer into the porous body proceeds through the body-porous mould boundary, so that the boundary condition $\boldsymbol{h} = -D_{\text{ef}}\, \delta_x C$ still holds. As a result of a high humidity of the environment, no liquid evaporates at the body-environment boundary, so that also the other boundary condition keeps its form $\delta_x C = 0$. On the basis of the given concept it follows that the boundary conditions introduced for body formation are analogous to the conditions of its solidification, and that the initial condition can be described by the expression

$$t = 0 \qquad x \in (0;L) \qquad c = c(x), \tag{7}$$

which, following rearrangement, has the form

$$j = 0 \qquad i \in (0,N) \qquad C_{i,j} = C(x,t) \tag{8}$$

The system of equations (3), (5), (8) allows the time development of liquid concentration in the body, and the mean concentration in the course of solidification to be calculated. If one knows the concentration of liquid in the body at which no more significant changes in volume take place, then it is possible to determine the time at which the body becomes separated from the porous mould, on the basis of the known time dependence of the mean concentration, or the time development of the concentration field in the body. The value of liquid concentration in the body at which the major changes in volume are concluded, can for instance be established from the dependence of body shrinkage on liquid phase concentration. The time of mechanical separation of the body from the mould then corresponds to the time when the mean concentration, or the body surface concentration at the body-mould boundary, has attained the given concentration value. After that time, no more liquid phase passes through the body-porous mould boundary and the body can be removed from the porous mould.

EXPERIMENTAL

In the case of body formation, the model allows the time development of the concentration profile, the mean concentration and the kinetics of body formation to be calculated. In the instance of body solidification, it permits the time development of the concentration profile, of the mean concentration, and the time of body solidification, to be determined. The model and the simplifying assumptions can be verified by comparing the results obtained from the model to those obtained experimentally. For a proper comparison, the experimental arrangement must correspond to the conditions for which the model has been prepared and the given relationships calculated In the given case, the following conditions have to be met:

- unidirectional transmission of the liquid phase through the system;
- determination of the initial conditions, i.e. distribution of the liquid phase at time t = O;
- determination of the boundary conditions, i.e. the time dependence of the surface flow of the liquid pha se, and ensuring zero flow through the other boundary of the system.

The verification of the system then consists of comparing the calculated values with the experimentally established ones, namely the time development of concentration profiles in the body, the mean concentration, the body thickness and the time of body solidification.

The experimental verification of the body formation and solidification model was implemented on casting a plate-shaped body. The measurements were carried out on an aqueous suspension of $\alpha - Al_2O_3$ at concentration $C = 0.5263$ m^3 m^{-3}.

The requirements specified above were ensured by the following experimental arrangement. The electrolyte employed and the way the suspension has prepared ensured homogenous distribution of the solid phase throughout the volume. The unidirectional character of transmission was provided by the method of pouring the suspension onto a gypsum pad, using the arrangement shown in Fig. 2.

The time dependence of the surface flow of liquid at the suspension-gypsum or suspension-body boundary was established by measuring the changes in the volume of the suspension. Maintaining a constant area

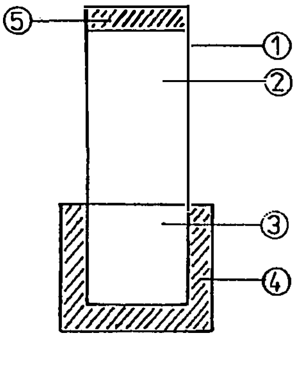

Fig. 2. Unidirectional plate-shaped body casting. 1 – graduated cylinder, 2 – suspension, 3 – gypsum pad, 4 – moisture barrier, 5 – closure.

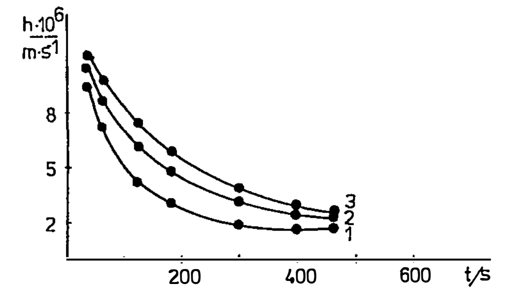

Fig. 3. Time dependence of liquid flow. 1 – unidirectional flow, 2, 3 – multidirectional flow in the gypsum.

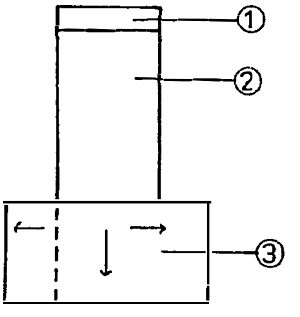

Fig. 4. Schematic diagram of multidirectional liquid flow through the gypsum mould. 1 – graduated cylinder, 2 – suspension, 3 – porous mould.

of the suspension or body and gypsum at the body-gypsum boundary, and moisture-proofing of the gypsum pad according to Fig. 4, proved to be significant factors ensuring a unidirectional flow of the liquid. In the given arrangement, it was possible to measure in a reproducible way the relationships $\boldsymbol{h} = \boldsymbol{h}(t)$ which showed the course plotted in Fig. 3, curve *1*. In the case when the area of gypsum at the body-gypsum boundary was larger than the area of the body and the gypsum pad was not moisture-proofed, quite different and irreproducible courses of the $\boldsymbol{h} = \boldsymbol{h}(t)$ relationship were obtained (Fig. 3, curves *2* and *3*). The difference in the results of the two procedures can be explained by failure to comply with the requirement for unidirectional flow of the liquid through the porous mould, as illustrated by Fig. 4.

In the case of body solidification, the time dependence of surface flow at the body-gypsum boundary was evaluated from the experimentally established time dependence of mean body concentration, shown in Fig. 5.

The saturation of the environment with water above the suspension level has ruled out vaporization of the liquid at this boundary. The time development of the concentration profile was determined by

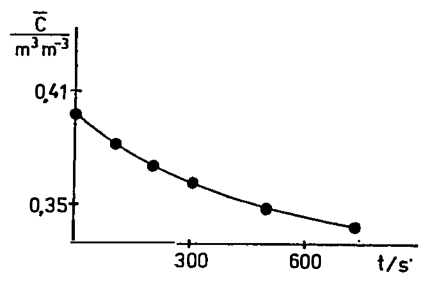

Fig. 5. Time dependence of mean concentration for body solidification.

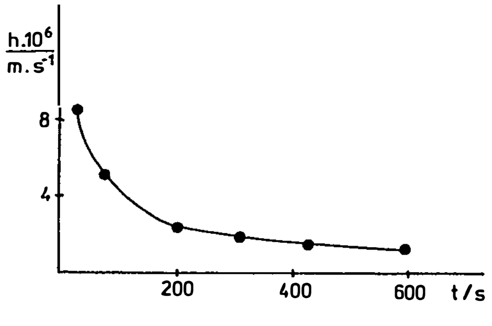

Fig. 6. Time dependence of liquid surface flow for body formation.

drying the samples, obtained by slicing the body, at various time intervals. The time dependence of body thickness and the mean concentration was determined by measuring the body thickness and the mean concentration at various time intervals. The experimental dependences of surface flow, body thickness and mean concentration for body formation are shown in Figs. 6, 7 and 8. For body solidification, the time relationships of mean concentration and surface flow are listed in Table I. The time development of concentration profiles in the body during its formation and solidification is given in Figs. 10 and 11.

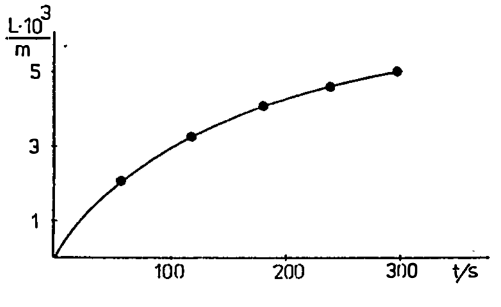

Fig. 7. Time dependence of body thickness.

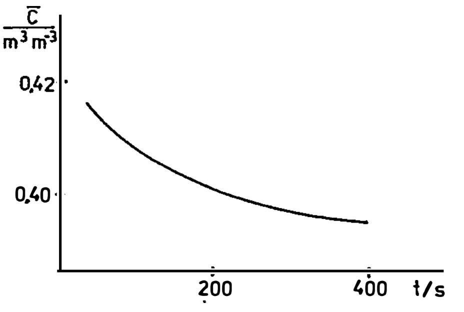

Fig. 8. Time dependence of mean concentration for body formation.

Verifying the model of body formation and solidification

The relationships seeked can be calculated from the mathematical model either by arbitrary choice of initial and boundary conditions, or by using experimentally established values. In the present study, the calculation was performed for experimentally determined boundary conditions (Figs. 6 and 9) and for the following values [2–5]:

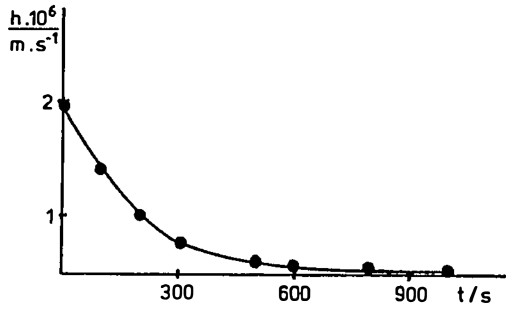

Fig. 9. Time dependence of liquid surface flow for body solidification.

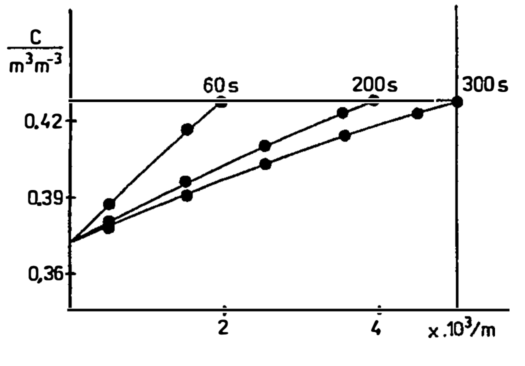

Fig. 10. Time dependence of concentration profile for body formation.

$C_0 = 0.53$ m^{3}m^{-3}; $C_k = 0.425$ m^{3}m^{-3};
$C_z = 0.3384$ m^{3}m^{-3};
$D = 2.45\times10^{-8}$ m^{2}s^{-1};
$D_p = 3.00\times10^{-13}$ m^{2}s^{-1}Pa^{-1};
$D_{ef} = 2.00\times10^{-7}$ m^2 s^{-1}.

Substitution of the values into the model and calculation yielded the relationships plotted in Figs. 12, 13, 14, 15 and 16. The times of body solidification corresponding to attainment of concentration C_z for various body thicknesses, are listed in Table I jointly with the experimental values for the sake of comparison.

CONCLUSION

The study was aimed at creating a mathematical model describing the formation and solidification of green ceramic body during the pouring of suspensions into porous moulds (the slip-casting process). In deriving the model, various simplifying assumptions were adopted. Suitability of a proposed model can only be verified by comparing the results obtained by calculations with those of experimentally established relationships. This comparison is viable solely when all the requirements introduced in deriving the model are conformed to during the experimental verification. In the given case, there are the following conditions in addition to the preparation of porous moulds with defined properties:

1. Reproducible preparation of the suspension with initial homogeneous distribution of the liquid,
2. Determination of the surface flow of liquid during body formation and solidification at the boundary between the porous mould and the suspension, as well as between the porous mould and the body,
3. Securing a zero flow at the boundary between the suspension and the environment, and between the body and the environment.

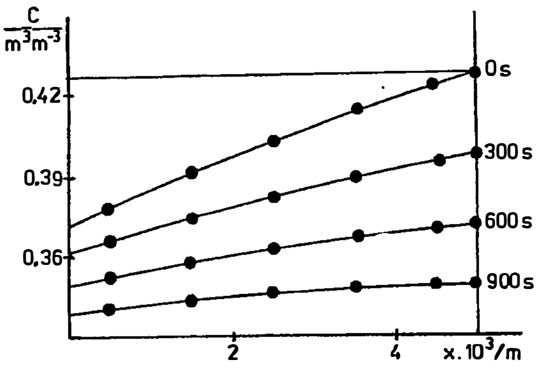

Fig. 11. Time dependence of concentration profile for body solidification.

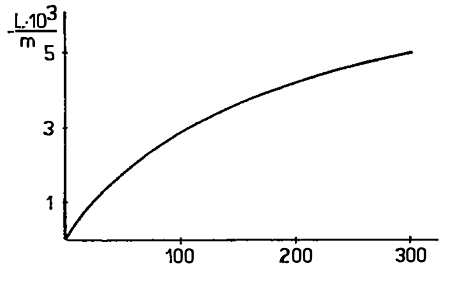

Fig. 12. Time dependence of body thickness – calculated.

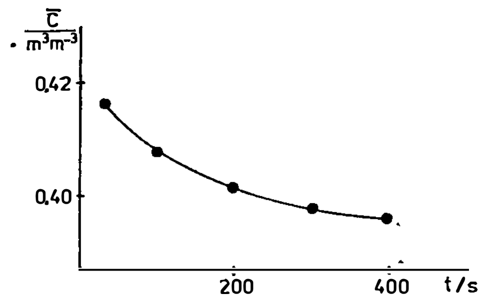

Fig. 13. Time dependence of concentration profile for body formation – calculated.

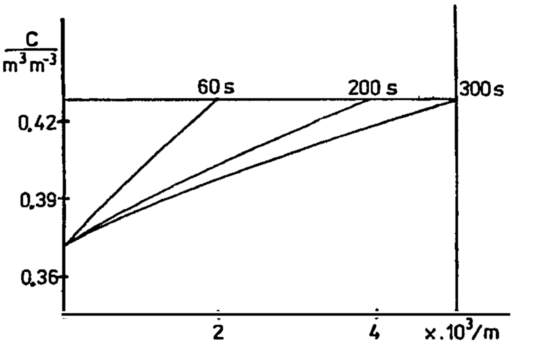

Fig. 14. Time dependence of concentration profile for body formation – calculated.

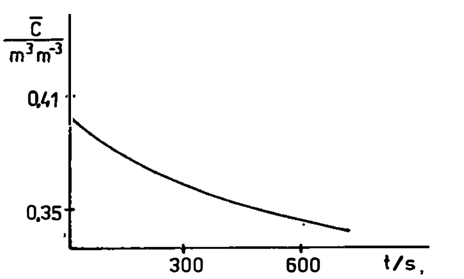

Fig. 15. Time dependence of mean concentration for body formation – calculated.

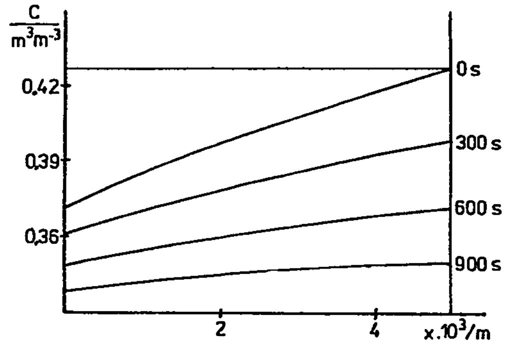

Fig. 16. Time dependence of concentration profile for body solidification – calculated.

Table I

Time of body solidification for various thicknesses

L [m]	t (calculated) [s]	t (measured) [s]
3×10^{-3}	830	874
3×10^{-3}	990	1053
7×10^{-3}	1220	1296

A comparison of the time dependences of concentration profiles, medium concentration, body thickness calculated from the time of solidification using the model (cf. Figs. 12, 13, 14, 15 and 16) with the relationships established experimentally (cf. Figs. 5, 6, 7, 8 and 9 and Table I) shows a satisfactory mutual agreement. The error evaluated by comparing the courses of the relationships does not exceed 9%.

The results obtained allow to reach the conclusion that the model of body formation and solidification describing the processes as diffusion in a binary mixture of incompressible components permits the course of body formation and solidification during slip casting (pouring of suspensions into porous moulds) to be predicted with a satisfactory accuracy.

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Translated by K. Němeček

MODEL LITÍ KERAMIKY

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Aplikací zobecněného difúzního přístupu je zavedena představa, že přenos kapalné fáze v technologii lití je vyvolán koncentrační difúzí a barodifúzí. Barodifúze souvisí s kapilárním sáním kapaliny v meniscích na nenasyceném rozhraní suspenze – porézní forma resp. střep – porézní forma. Na základě této představy byl vypracován matematický model tvorby a zatuhování střepu pro výpočet časového vývoje koncentračních polí, střední koncentrace, kinetiky tvorby střepu a doby zatuhnutí střepu.

Ověření matematického modelu tvorby střepu bylo provedeno pro vodnou suspenzi α-Al_2O_3 porovnáním časových závislostí koncentračních profilů, střední koncentrace a tloušťky střepu vypočtených z modelu s experimentálně stanovenými závislostmi. Model tvorby střepu fenomenologicky popisující operaci jako difúzi v binární směsi nestlačitelných složek umožňuje předpovídat průběh tvorby střepu s chybou menší než 9%.

Obr. 1. Schématické znázornění tvorby střepu.

Obr. 2. Jednosměrné lití desky. 1–odměrný válec, 2–suspence, 3–sádrová podložka, 4–vlhkostní izolace, 5–uzávěr.

Obr. 3. Závislost toku kapaliny na čase. 1–jednosměrný tok; 2,3–vícesměrný tok v sádře.

Obr. 4. Schéma vícesměrného přenosu kapaliny porézní formou. 1–odměrný válec, 2–suspenze, 3–porézní forma.

Obr. 5. Časová závislost střední koncentrace pro zatuhování střepu.

Obr. 6 Časová závislost střední koncentrace pro zatuhování střepu.

Obr. 7. Časová závislost tloušťky střepu.

Obr. 8. Časová závislost střední koncentrace pro tvorbu střepu.

Obr. 9. Časová závislost povrchového toku kapaliny pro zatuhování střepu.

Obr. 10. Časová závislost koncentračního profilu pro tvorbu střepu.

Obr. 11. Časová závislost koncentračního profilu pro zatuhování střepu.

Obr. 12. Časová závislost tloušťky střepu – vypočtená.

Obr. 13. Časová závislost střední koncentrace pro tvorbu střepu – vypočtená.

Obr. 14. Časová závislost koncentračního profilu pro tvorbu střepu – vypočtená.

Obr. 15. Časová závislost střední koncentrace pro zatuhování střepu – vypočtená.

Obr. 16. Časová závislost koncentračního profilu pro zatuhování střepu – vypočtená.