# METHOD FOR DETERMINING THE EFFECTIVE DIFFUSION COEFFICIENT OF LIQUID IN THE SYSTEM MOULD – BODY – SUSPENSION

MIROSLAV KOVÁČ, JIŘÍ HAVRDA

Institute of Chemical Technology, Department of Glass and Ceramics, Technická 5, 168 28 Prague 6

Received 27. 5. 1991

The transfer of liquid in the system porous mould – body was characterized by the effective diffusion coefficient. A method has been developed for determining the coefficient, and a procedure for calculating its value from experimentally established time dependence of the mean concentration and the thickness of the body formed.

#### INTRODUCTION

Transfer of liquid through a saturated ceramic mix is affected by the state of the ceramic mixenvironment boundary [1]. If the boundary is unsaturated by the liquid, its transfer due to a concentration gradient is speeded up by capillary suction in the menisci at this boundary, i.e. by barodiffusion. Such a situation occurs e.g. in the course of drying of ceramic bodies [2]. In the given case a diffusion coefficient expressed solely on the basis of concentration diffusion has the significance of the effective diffusion coefficient which also includes liquid transfer by capillary barodiffusion. An unsaturated boundary, similarly to drying of ceramic bodies, also arises in the casting of slips or suspensions into porous moulds, namely at the boundary of the suspension, the body porous mould boundary. Determination of the effective diffusion coefficient of a liquid in a system with an unsaturated boundary in the course of body formation is dealt with in the present study.

## THEORETICAL PRINCIPLES OF THE DETERMINATION OF THE EFFECTIVE DIFFUSION COEFFICIENT

In a system with an unsaturated boundary, i.e. a system where the transfer of liquid in a mix is accelerated by capillary suction in the menisci at the unsaturated boundary, the effective diffusion coefficient involved in the transfer of liquid through the ceramic body during its formation and solidification, can be determined by the method based on measuring the time dependence of the liquid flow, namely by pouring the suspension onto a porous plate and examining the flow of the liquid into the pad by measuring the speed at which the suspension level falls, the mean concentration and the thickness of the forming body.

If the liquid transfer during body formation is unidirectional, the mass balance has the form

$$\delta_t C = D_{\rm ef} \, \delta_{xx} C \tag{1}$$

and for the volume flow of the liquid it holds that

$$\boldsymbol{h} = -D_{\rm ef} \,\delta_x \boldsymbol{C} \tag{2}$$

Numerical solution of equation (1) by the grid method using step h and time step k for the conditions

$$\begin{array}{ll} t = 0 & x \in \langle 0; H \rangle & C = C_0 & (3) \\ t > 0 & x = 0 & h = h(t) \\ x = L(t) & C = C_k & (4) \end{array}$$

is used to express the equation for calculating the mean concentration C:

$$\overline{C} = L^{-1}((C_i + C_{i-1})) h/2$$
(5)

and for the kinetics of body formation one obtains

$$L = (6)$$
  
(C<sub>k</sub> - C<sub>i-1</sub>)(X<sub>i</sub> - X<sub>i-1</sub>)/(C<sub>i</sub> - C<sub>i-1</sub>) + X(i - 1)

where H is the height of the suspension column, L is body thickness, t is time, C is concentration by volume, X is the ordinate and  $C_k$  is the coagulating concentration.

On choosing  $D_{ef}$  and comparing the calculated time dependences C(t) and L(t) according to equations (5) and (6) with the courses established experimentally, one can evaluate the  $D_{ef}$  being seeked when the relationships show mutual agreement.

### EXPERIMENTAL RESULTS

The effective diffusion coefficient was determined for an aqueous suspension of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> with a particle size over the interval of  $(0.1\mu m; 0.4\mu m)$ . Following addition of a suitable electrolyte, the content of solids in the suspension was in the range of  $(0.43 \text{ m}^3\text{m}^{-3} \cdot$  $0.66 \text{ m}^3\text{m}^{-3})$ .

The experimental arrangement for measuring the effective diffusion coefficient is shown in Fig. 1.

The unidirectional transfer of the liquid during the measurement was ensured by maintaining an identical surface area of the suspension and the gypsum pad at the boundary, and by waterproofing the gypsum pad. The arrangement allowed the flow of the liquid to be measured at the same time with measuring the thickness of the arising body. The samples for medium moisture content determination were taken from the newly formed body at times t.



Fig. 1. Schematic diagram of the experimental arrangement for measuring  $D_{ef}$ . 1 – measuring cylinder, 2 – gypsum pad, 3 – waterproofing insulation, 4 – micrometric screw, 5 – suspension

Typical experimental time dependences of the liquid flow, the body thickness and the mean concentration are plotted in Figs. 2, 3 and 4 for a suspension with a solids content of  $0.4637 \text{m}^3\text{m}^{-3}$ . For the sake of comparison, Figs. 3 and 4 also show the calculated courses for a selected  $D_{\text{ef}}$ . Similar relationships were also measured for other suspensions having solids content over the interval  $(0.4466 \text{m}^3\text{m}^{-3}; 0.6496 \text{m}^3\text{m}^{-3})$ . A survey of the  $D_{\text{ef}}$  evaluated is listed in Table I.

 Table I

 Values of effective diffusion coefficients

<i>C</i> ₀ [m <sup>3</sup> m <sup>-3</sup> ]	$D_{ef}$ $[m^2s^{-1}]$	$D_{ef}$ .[m²s <sup>-1</sup> ]
0.6496 0.6051 0.5264 0.4637	$2.0 \times 10^{-7}$ $2.0 \times 10^{-7}$ $2.1 \times 10^{-7}$ $1.9 \times 10^{-7}$	2.0×10 <sup>-7</sup>

The results indicate that the method allowed the effective diffusion coefficient to be reliably determined and that its value was  $D_{\rm ef} = 2 \times 10^{-7} {\rm m}^2 {\rm s}^{-1}$ .

The method for determining the effective diffusion coefficient was also utilized for determining this coefficient for the liquid in the porous mould. In that case, the system consists solely of pure liquid and the porous mould, and no body formation takes place in the system. The time dependence of the liquid flow at the liquid – porous mould boundary, the mean concentration of the liquid, and its distribution in the porous mould were measured. The concentration profile of the liquid in the porous mould was established by drying the samples obtained by cutting the mould into slices at a given time. The chosen  $D_{\rm ef}$ , corres-



Fig. 2. Time dependence of liquid flow.



Fig. 3. Time dependence of body thickness. 1 – experimental, 2 – calculated



Fig. 4. Time dependence of the mean body concentration. 1 - experimental, 2 - calculated

ponding to an agreement between the calculated and the experimentally determined course of mean concentration and concentration profile in the porous mould, is regarded as the effective diffusion coefficient being seeked.

Typical experimentally established relations h = h(t),  $\overline{C} = \overline{C}(t)$  and C = C(x, t) are shown in Figs. 5, 6 and 7.

#### CONCLUSION

The method described above allowed the effective diffusion coefficient of the liquid,  $D_{ef}$ , involved in the body formation from an  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> suspension, to be reliably determined. The results indicate that over



Fig. 5. Time dependence of the liquid surface flow.



Fig. 6. Time dependence of mean concentration.



Fig. 7. Concentration profile of the liquid in gypsum. 1 - t = 60 s, 2 - t = 180 s, 3 - t = 300 s

the interval  $C \in \langle 0.43 \text{m}^3 \text{m}^{-3}; 0.66 \text{m}^3 \text{m}^{-3} \rangle$ , the  $D_{\text{ef}}$  was independent of concentration and its value was  $D_{\text{ef}} = 2 \times 10^{-7} \text{m}^2 \text{s}^{-1}$ . The effective diffusion coefficient including the transfer of liquid by concentration diffusion and barodiffusion was higher by one order of magnitude than the diffusion coefficient of purely

concentration diffusion  $(D = 2.45 \times 10^{-8} \text{m}^2 \text{s}^{-1})$  [3].

The results have further shown that the effective diffusion coefficient of liquid in a gypsum mould can also be regarded as being independent of concentration, and its value was  $D_{\rm ef} = 2.72 \times 10^{-7} {\rm m}^2 {\rm s}^{-1}$ .

#### References

- [1] Havrda J., Oujiří F.: Silikáty 26, 107 (1982).
- [2] Havrda J., Oujiří F., Žižková M.: Silikáty 28, 1 (1984).
- [3] Kováč M., Havrda J.: Ceramics-Silikáty (1992), in press.

Translated by K. Němeček

### METODA STANOVENÍ EFEKTIVNÍHO DIFÚZNÍHO KOEFICIENTU KAPALINY V SYSTÉMU FORMA-STŘEP-SUSPENZE

Miroslav Kováč, Jiří Havrda

### Vysoká škola chemicko-technologická, Ústav skla a keramiky, Technická 5, 166 28 Praha 6

Pro popis technologie lití keramických suspenzí je nutná znalost difúzních charakteristik zpracovávaných směsí. Z hlediska popisu je možno rozlišit dva přístupy řešení. První přístup neuvažuje vliv stavu rozhraní a stanovené charakteristiky jsou charakteristiky efektivní. Druhý přístup uvažuje vliv stavu rozhraní a stanovené hodnoty považuje za skutečné difúzní charakteristiky.

Pro stanovení efektivního difúzního koeficientu lze aplikovat metodu založenou na výpočtu jeho hodnoty z experimentálně stanovených hodnot střední koncentrace a tloušťky vzniklého střepu. Tato metoda byla aplikována i na stanovení difúze kapaliny nasycenou keramickou směsí oxidu hlinitého a stanovená hodnota efektivního difúzního koeficientu byla  $2.10^{-7}$  m<sup>-2</sup>s<sup>-1</sup>.

- Obr. 1. Schéma uspořádání metody měření Dei.
  - 1 odměrný válec, 2 sádrová podložka,
  - 3 vlhkostní izolace, 4 mikrometrický šroub,
    5 suspenze
- Obr. 2. Časová závislost toku kapaliny.
- Obr. 3. Časová závislost tloušťky střepu.
- 1 namērená, 2 vypočtená
- Obr. 4. Časová závislost střední koncentrace střepu. 1 – naměřená, 2 – vypočtená
- Obr. 5. Závislost povrchového toku kapaliny na čase.
- Obr. 6. Závislost střední koncentrace na čase.
- Obr. 7. Koncentrační profil kapaliny v sádře.
  - 1 t = 60 s, 2 t = 180 s, 3 t = 300 s