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Numerical Analysis of Defects Caused by Thermolysis in an Infinite Cylindrical Ceramic Moulding

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ABSTRAK

Aturan-kedua persamaan parabolik (resapan) yang menentukan kepekatan monomer semasa pirolisis pembentukan silinder seramik, mengandungi polimer yang merendahkan monomer sahaja diselesaikan: silinder yang tidak boleh diukur diberi perhatian. Kaedah garisan digunakan untuk mengubah sebahagian persamaan pembezaan kepada sistem linear aturan-pertama persamaan pembezaan biasa, suatu penyelesaian yang terbukti dapat meyakinkan hubungan berulangan. Profil kepekatan pada tempoh yang diberikan ditaksir dengan menggantikan istilah-istilah eksponen matrik dalam hubungan berulangan dengan taksiran Padé aturan-tinggi. Algoritma selari menggunakan dua pemproses, dibangunkan dengan mengambil sebahagian pecahan penguraian anggaran Padé dan gambaran rasional (matriks) dalam hubungan berulangan.

ABSTRACT

The second-order parabolic (diffusion) equation, which governs the concentration of monomer during pyrolysis of a cylindrical ceramic moulding containing a polymer which degrades to monomer only, is solved: an infinite cylinder is considered. The method of lines is used to transform the partial differential equation into a system of first-order linear ordinary differential equations, the solution of which is seen to satisfy a recurrence relation. The concentration profile at a given time is computed by replacing the matrix exponential terms in this recurrence relation by a high-order Padé approximant. A parallel algorithm using two processors is developed by taking the partial-fraction decomposition of the Padé approximant and a (matrix) rational expression in the recurrence relation.

Keywords: numerical analysis defects, thermolysis, infinite cylindrical ceramic moulding

INTRODUCTION

The solution of the transport problem in geometries such as the cylinder is important to the technologically significant problem of forming of engineering ceramics. Manufacturing techniques resulting from polymer processing can now be applied to the fabrication of engineering ceramic components by a powder route. The most versatile of such techniques is injection moulding (Edirisinghe and Evans 1986a, b). These processes incorporate the powder in

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an organic vehicle, preferably a polymer, in order to effect shaping. The organic matter is then removed, usually by controlled thermolysis, before sintering the powder assembly. Attempts have been made to quantify the thermolysis stage, and these have been reviewed by Shaw and Edirisinghe (1995).

Calvert and Cima (1990) and Evans *et al.* (1991) addressed the critical mechanism of thermal degradation of the organic phase at an early stage of the pyrolysis when all pores are filled. Mass transport occurs by diffusion of degradation product in the parent polymer which forms the continuous phase in the crowded two-phase composite at the critical stage. These models predict the incidence of defects by applying the criterion of boiling to the solution of the degradation product in the parent polymer. Matar *et al.* (1993) and Shaw and Edirisinghe (1995) extended the modelling to consider the effect of porosity development on mass transport.

Experimentally, the determination of the critical heating rate, which is the fastest linear heating rate resulting in a defect-free thermolysis product, is laborious as it involves thermolysis at different rates for a large number of samples for each section size. In the present paper the heat transfer (diffusion) equation for an infinite cylinder will be solved using a parallel algorithm. The variation of vapour pressure of monomer over solution during a linear temperature ramp is determined by finding the centre concentration of monomer in the cylinder. A bubble forms when the vapour pressure exceeds the ambient pressure, and if this happens the ceramic component will be defective. However, if the rate of heating is sufficiently low to prevent the vapour pressure rising to ambience, the ceramic will be free from related defects.

THE MATHEMATICAL MODEL

The problem modelled is that of ceramic fabrication in which the decomposition of polymer yields an internal source of monomer only. Monomer evaporates from the surface and hence a concentration profile develops. The objective is to find the centre concentration and apply the criterion for boiling.

An infinite cylinder of radius B, injection-moulded using ceramic-polymer suspension, is uniformly heated along its length at a constant rate Z Ks^{-1}. Assuming radial symmetry, the concentration profile of monomer, $c = c(r,t)$, throughout the cylinder at a given time t satisfies the partial differential equation (PDE)

$$\frac{\partial c}{\partial t} = D\left(\frac{1}{r}\frac{\partial c}{\partial r} + \frac{\partial^2 c}{\partial r^2}\right) + Q;\ 0 \le r < B,\ t > 0 \qquad (1)$$

(Evans et al. 1991) in which D is the diffusion coefficient of the material. In practice, $D = D(c, T)$, where T represents temperature at time t, and can vary by a factor of 105, but for the purpose of the analysis its (updated) *value* at time t will be used (effectively treating D as a constant at time t). Such a localized value of a variable is often referred to by numerical analysts as the *frozen* value of the variable. Clearly, symmetry requires this PDE to be solved only in the half interval $0 \leq r \leq B$. This, in turn, imposes the flux boundary condition

$$\frac{\partial c(0,t)}{\partial r} = 0, \ t > 0 \ ; \ r = 0 \tag{2}$$

on the axis of the cylinder and the assumption of zero concentration on the surface gives rise to the boundary condition

$$c(B, T) = 0, \ t > 0 \tag{3}$$

Initially it will be assumed that the concentration throughout the cylinder is $c^* \neq 0$, giving rise to the intitial condition

$$c(r,0) = C^*, \ 0 \leq r \leq B \tag{4}$$

Note, there is a discontinuity between initial and boundry conditions.

In (1), $\dot{Q}$ is the rate of production of monomer caused by pyrolysis based on the total volume of ceramic-powder suspension and is given by

$$\dot{Q} = K_o \exp\left[-E/(RT)\right] \rho_p (1 - V_c) H \tag{5}$$

(Evans *et al.* 1991), where H is the mass fraction of polymer remaining after a given absolute temperature is reached and is given by

$$H = \exp\left[-\frac{K_o R T^2 \exp\left[-E/(RT)\right]}{ZE}\left\{1 - \frac{2RT}{E} + \frac{6(RT^2)}{E^2}\right\}\right] \tag{6}$$

The parameters in (5) and (6) are described in the nomenclature. Clearly, $\dot{Q}$, which is a function of T, is not constant but at a given time t its (frozen) value at that time will be used in the algorithm.

The PDE (1) cannot be solved on the axis of the cylinder (r=0). Using the boundary condition (2) in the Maclaurin expansion

$$\frac{\partial c(r,t)}{\partial r} = \frac{\partial c(0,t)}{\partial r} + \frac{\partial^2 c(0,t)}{\partial r^2} + \frac{r^2}{2}\frac{\partial^3 c(0,t)}{\partial r^3} + \ldots \tag{7}$$

it is easily seen that

$$\lim_{r \to 0} \frac{1}{r}\frac{\partial c(r,t)}{\partial r} = \frac{\partial^2 c(0,t)}{\partial r^2} \tag{8}$$

so that the alternative PDE

$$\frac{\partial c}{\partial t} = 2D\,\frac{\partial^2 c}{\partial r^2} + \dot{Q} \tag{9}$$

must be solved when r = 0.

THE NUMERICAL SOLUTION

Discretization and the Method of Lines

The space interval $0 \leq r \leq B$ is subdivided into M + 1 parts each of width h so that $(M + 1)h = B$ and the time variable $t \geq 0$ is incremented in steps of length l so that t may be wirtten in the form $t = t_n = n\,l$ where n is an integer. The region Ω in (r,t) space in which the concentration is to be computed and its boundary $\partial\Omega$ consisting of the lines r = 0, r = a and t = 0 have thus been covered by a grid of points $(r_m, t_n) = (mh, n\,l)$ where m = 0, 1, ... M, M + 1 and n = 0, 1, 2, ...

The space derivative in (20) is approximated by the second-order central-difference replacement

$$\frac{\partial c(r,t)}{\partial r} = (2h)^{-1}\big[c(r + h,t) - c(r - h,t)\big] + 0(h^2) \text{ as } h \rightarrow 0 \tag{10}$$

with $r = r_0 = 0$, to give

$$C^n_{-1} = C^n_1 + 0(h^3) \text{ as } h \rightarrow 0 \tag{11}$$

in which upper case C is used to distinguish the solution of differential equation. Using this notation, C^n_m is an approximation to the concentration $c(r_m, t_n) = c(mh, n\,l)$ with m = 0, 1, ..., M, M + 1 and n = 0, 1, 2, ... Thus m = 0 refers to points on the axis of the cylinder, m = M + 1 to points on the surface of the cylinder and n = 0 to the initial distribution. The quantity C^n_{-1} appears to be outside $\Omega \cup \partial\Omega$ but this term will be required in the algorithm. It will be convenient to use the vector $C^n = \left[C^n_0, C^n_1, \ldots, C^n_m\right]^T$, T denoting transpose.

The space derivative in (9) will be replaced by

$$\frac{\partial^2 c(r,t)}{\partial r^2} = h^{-2}\big[c(r + h,t) - 2c(r,t) + c(r + h,t)\big] + 0(h^2) \text{ as } h \rightarrow 0 \tag{12}$$

wih $r = r_0$ and those in (1) by (10) and (12) with $r = r_m$ (m = 1, 2, ..., M). Then, applying (9) with (12) and (11) to the point $r = r_0$ and (1) with (10) and (12) to the points $r = r_m$ $(m = 1, 2, \ldots, M)$ at time $t = tn = n\,l$ leads to the (semi-discrete) linear, initial-value problem (IVP) given by

$$\frac{dC(t)}{dt} = AC(t) + q_t,\ t > 0,\ C(0) = g \tag{13}$$

in which $q_t = \left[\dot{Q}_t, \ldots, \dot{Q}_t\right]^T$, $g = \left[C^*, \ldots C^*\right]^T$ are vectors of order $M + 1$ with $\dot{Q}_t$ denoting the (frozen) value of $\dot{Q}$ at time t (see §2). It is appropriate to treat the vector q_t as a constant vector at any time t because $\dot{Q}$, given in equation (5), is not a function of t. The temperature, T, in (5) is a measured value and is not a known function of t. In (13) A is the tridiagonal matrix of order M + 1 given by

$$A = Dh^{-2}\begin{bmatrix} -4 & 4 & & & \\ \alpha_1 & -2 & \beta_1 & & \\ & & \ddots & & \\ & & \alpha_{m-1} & -2 & \alpha_{m-1} \\ & & & \alpha_m & -2 \end{bmatrix} \tag{14}$$

with $\alpha_m = 1 - 1/(2m)$ for $m = 1,2, \ldots, M$ and $\beta_m = 1 + 1/(2m)$ for $m = 1,2,\ldots, M-1$. It may be shown using Brauer's theorem (Gerschgorin's circle theorem) that the eigenvalues of matrix A have negative real parts (Twizell 1984).

A Recurrence Relation

Treating $q_t = q$ as a constant vector at time t, the solution of the IVP (13) is given by

$$C(t) = -A^{-1}q + A^{-1}\exp(tA)(q + A^{-1}g) \tag{15}$$

Which may be shown to satisfy the recurrence relation

$$C(t + l) = -A^{-1}q + \exp(lA)[C(t) + A^{-1}q] \tag{16}$$

It is this recurrence relation which may be used to develop a numerical method for the solution of (13) and thus of (9), (1-4). Numerical methods may be derived by making approximations to exp (lA) in (16). The reader is referred to the papers by Lawson (1972), Lawson and Swayne (1976), Swayne (1987, 1988), Twizell *et al.* (1996) and Taj and Twizell (1997) for a recurrence relation which may be used when the vector **q** is a function of t.

A Parallel Algorithm

Approximating the exponential term in (16) by its (4, 0) Padé approximant gives

$$C(t+l) = -A^{-1}q + \left(I - lA + \tfrac{1}{2}l^2A^2 - \tfrac{1}{6}l^3A^3 + \tfrac{1}{24}l^4A^4\right)^{-1}\left[C(t) + A^{-1}q\right] \quad (17)$$

The approximant is chosen because of its known high-order accuracy (Khaliq 1983). Pre-multiplying by $\left(I - lA + \tfrac{1}{2}l^2A^2 - \tfrac{1}{6}l^3A^3 + \tfrac{1}{24}l^4A^4\right)$ and rearranging gives

$$\begin{aligned}&\left(I - lA + \tfrac{1}{2}l^2A^2 - \tfrac{1}{6}l^3A^3 + \tfrac{1}{24}l^4A^4\right)C(t+l)\\ &= C(t) + \left(I - \tfrac{1}{2}lA^2 + \tfrac{1}{6}l^2A^2 + \tfrac{1}{24}l^3A^3\right)\end{aligned} \quad (18)$$

It is easy to show that the order of (18) is $0(h^2 + l^4)$ as $h, l \to 0$ and that, when $q \equiv 0$, the method is L-stable.

To achieve parallel implementation, (18) must first be written in the form

$$\begin{aligned}C(t+l) &= \left(I - lA + \tfrac{1}{2}l^2A^2 - \tfrac{1}{6}l^3A^3 + \tfrac{1}{24}l^4A^4\right)^{-1}C(t)\\ &+ \left(I - lA + \tfrac{1}{2}l^2A^2 - \tfrac{1}{6}l^3A^3 + \tfrac{1}{24}l^4A^4\right)^{-1}\left(I - \tfrac{1}{2}lA - \tfrac{1}{6}l^2A^2 + \tfrac{1}{24}l^3A^3\right)lq\end{aligned} \quad (19)$$

following which partial-fraction decomposition of the (4, 0) Padé approximant to exp (lA) itself and of the matrix rational expression (the coefficient of lq) in (19) gives

$$C(t+l) = \sum_{j=1}^{4}\left[s_j(I - b_j lA)^{-1}C(t) + k_j(I - b_j lA)^{-1}lq\right], \quad (20)$$

in which

$$b_1 = \bar{b}_2 = 0.426266565027023\ 10^{-1} + i\ 0.39463295317211,$$

$$b_3 = \bar{b}_4 = 0.45737334349730 + i\ 0.23510048799854,$$

$$s_i = \bar{s}_2 = -0.12116960248677 - i\ 0.20306415938099,$$

$$s_3 = \bar{s}_4 = 0.62116960248677 - i\ 0.59941529409522,$$

$$k_1 = \bar{k}_2 = -0.516505502377253\ 10^{-2} - i\ 0.865594616994373\ 10^{-2},$$

$$k_3 = \bar{k}_4 = 0.28410641796826 - i\ 0.27415657720375.$$

Closed-form expressions for the complex constants b_j, s_j, k_j ($j = 1, 2, 3, 4$) are not available.

The solution vector $c(t + l)$ may then be comuted using the parallel algorithm

Processor 1: $(I - r_1 lA)W_1 = s_1C(t)$,

$(I - r_1 lA)W_2 = k_1 lq$;

Processor 2: $(I - r^3 lA)W_3 = s_3C(t)$

$(I - r3 lA)W4 = k3C(t)$

Then: $C(t + l) = 2[Re(W_1) + Re(W_2) + Re(W_3) + Re(W_4)]$

$q: = \left[\dot{Q}_{t+1},..,\dot{Q}_{t+1}\right]^T$, $t: = t + l$

in which both coefficient matrices are tridiagonal.

The once-only decompositions of the tridiagonal matrices $I - r_1 lA$ and $I - r_3 lA$ are carried out by processors 1 and 2, respectively. Each processor employs a tridiagonal solver based on LU-decomposition employing forward and backward substitution, to compute the intermediate vector W_j $(j = 1,2,3,4)$ at every time step.

This parallel algorithm is to be preferred to the sequential algorithm based on (18) which requires higher powers of the matrix A, because the increased band-width of the coefficient matrix $I - lA + \frac{1}{2} l^2A^2 - \frac{1}{6}l^3A^3 + \frac{1}{24}l^4A^4$ increases computing costs and storage requirements. An alternative $0(h^2 + l^4)$ method which uses real arithmetic and four processors, and is also L-stable, was developed by Voss and Khaliq (1996) and could be adapted for solving (1) - (4). Arguably the best-known, single-processor method which could be adapted for solving (1)-(4) is the Crank-Nicolson method (Crank and Nicolson 1947). Unfortunately, this method is only $0(h^2 + l^2)$ accurate as $h, l \to 0$ and is only A-stable, whereas the method given by (18) is L-stable. Lawson and Morris (1978) gave an excellent description of the short-comings of the Crank-Nicolson method when used to solve problems which have a discontinuity between the boundary condition (3) and the initial condition (4). Lambert (1991), for instance, lists the properties of approximations to $\exp(lA)$ in (16) which lead to A-stable or L-stable methods.

The problem solved in the present paper given by (1)-(4) is one-dimensional because of symmetry. Manufactured, ceramic artefacts usually have some symmetric properties which should always be exploited in making calculations. There will be occasions, however, when there will be a need to retain more than one space variable and in solving such problems parallelism in space should be taken into account. Lawson and Morris (1978) and Swayne (1987, 1988) developed single-processor algorithms for solving multi-dimensional PDEs while Twizell *et al.* (1996) and Taj and Twizell (1997) have developed multi-processor techniques for solving them.

BOILING CRITERION: NUMERICAL RESULTS

The vapour pressure at the base time level t_n is given by

$$P_v^n = \exp\left[-\Delta H_v/(RT^n) + a\right] \tag{21}$$

(Evans *et al.* 1991), in which Tn is the temperature at time tn, and this is updated at time t_{n+1} by the relation

$$P_v^{n+1} = P_v^n V_m \exp\left[1 - V_m + \chi(1 - V_m)^2\right] \tag{22}$$

Where

$$V_m = C_p P_1^{n+1} / \left[C_p V_1^{n+1} + (1 - C_p) V_2^{n+1}\right] \tag{23}$$

in which

$$V_1^{n+1} = 0.001098\left[1 + 0.0009774\left\{(n + 1)1 + 100\right\}\right] \tag{24}$$

$$V_2^{n+1} = 0.0009595\left[1 + 0.0004985\left\{(n + 1)1 + 49\right\}\right] \tag{25}$$

and

$$C_p = C_0^{n+1} / \left\{\rho_p(1 - V_c)\right\} \tag{26}$$

(Matar *et al.* 1993).

A bubble forms when the vapour pressure exceeds the atmospheric pressure. A plot of vapour pressure against temperature computed using the procedure described above with the data given in Table 1 is depicted in *Fig 1.* Unfortunately, no closed-form solution of the problem (9, 1-4) is available against which the

TABLE 1
Parameter values used for computation

Parameter	value	Uit
a	22.255	Pa
1	0.5	s
B	0.005	m
D	0.000692	m^2s^{-1}
E	222000	$J\ mol^{-1}$
ΔH_v	38940	$J\ mol^{-1}$
K_0	$1.67\ 3\ 10^{16}$	s^{-1}
M	99	–
R	8.3143	$J\ mol^{-1}K^{-1}$
V_c	0.5	dimensionless
ρ_p	1075	$kg\ m^{-3}$
χ	0.361	dimensionless

Nomenclature

Parameter	Description	Units
a	constant	Pa
b_j, k_j, s_j	constants in partial-fraction decmposition (j=1,2,3,4)	–
$\bar{b}_j$,$\bar{k}_j$ $\bar{s}_j$	complex conjugates of b_j, k_j, s_j (j = 1,2,3,4)	–
c	concentration of monomer, c = c(r,t)	kg m^{-3}
g	vector of order M, g = $[C^*, \ldots, C^*]^T$	–
h	increment in r	m
i	i = $+\sqrt{-1}$	–
l	increment in t	s
q_t, q	vectors of order M, qt = q = $\left[\dot{Q}_t,\ldots,\dot{Q}_t\right]^T$	–
r	space co-ordinate of point on radius of cylinder $(0 \le r \le B)$	m
t	time	s
A	square matrix of order M	–
B	radius of cylinder	m
C	numerical approximation to c	kg m^{-3}
C*	initial value of c	kg m^{-3}
D	diffusion coefficient	m^2s^{-1}
E	activation energy for thermal degradation	J mol^{-1}
H	mass fraction of polymer remaining	$0 < H < 1$
ΔH_v	enthalpy of vaporization	J mol^{-1}
I	identity matrix of order M	–
K_0	frequency factor for thermal degradation	s^{-1}
M	No. discretization points in $0 < r < B$	–
P_v	vapour pressure of monomer over the pure liquid	Pa
$\dot{Q}$	production rate of monomer caused by pyrolysis	kg $m^{-3}s^{-1}$
R	gas constant	J $mol^{-1}K^{-1}$
T	temperature	K
V_c	volume fraction of ceramic in ceramic-polymer body	$0< V_c <1$
V_m	volume fraction of monomer in polymer	$0< V_m <1$
V_1	specific volume of monomer	m^3kg^{-1}
V_2	specific bolume of monomer	m^3kg^{-1}
W_j	intermediate vecotrs (j = 1,2,3,4)	–
Z	heating rate	K s^{-1}
ρ_p	density of polymer	kg m^{-3}
χ	interaction parameter for monomer-polymer system	–
Ω	region in (r,t) plane	–
$\partial\Omega$	boundary of Ω	–
0	zero vector of order M	–
T	transpose of a vector	–

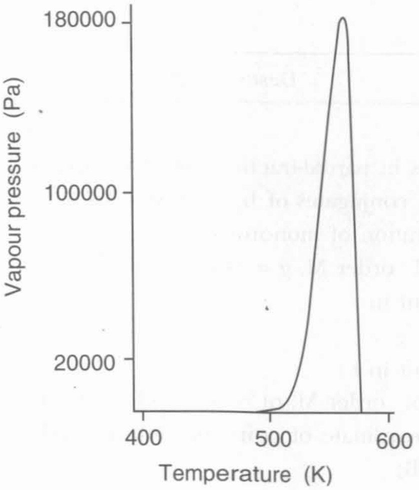

Fig 1. A plot of vapour pressure computed against temprature

profile in *Fig 1* could be compared, though the profile is in good agreement with other results in the literature (Matar *et al.* 1993). The parallel algorithm gave a speed-up figure of 1.02 and an efficiency figure of 51%; these figures are low because of the large number of operations which had to be carried out at each time step to update the quantities difined by (21)-(26)

CONCLUSION

A parallel algorithm has been developed for solving the second-order parabolic partial differential equation which governs the concentration of monomer during pyrolysis of a cylindrical ceramic moulding containing a polymer which degrades to monomer only.

The initial/boundary-value problem (IBVP) associated with the model was transformed into an initial value problem (IVP) in system form using the method of lines. The solution of the IVP was seen to satisfy a recurrence relation in which a matrix-exponential term was replaced by the (4,0) Padé approximant leading to an L-stable numerical method. The method was seen to be suitable for parallel implementation on an architecture with two processors. Comparisons were made with existing method from the literature.

In a numerical experiment the concentration profile was monitored at discrete points along the radius of a cylinder as the computation proceeded. The concentration on the central axis was used in the calculation of the vapour pressure, which is also known to depend on the temperature. A bubble forms when the vapour pressure exceeds the atmospheric pressure. No closed-form solution of the IBVP solved is available though the profile of vapour pressure against temperature, calculated using the parallel algorithm developed in the paper, was seen to be in good agreement with results appearing in the literature.

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