JOURNAL OF MATERIALS SCIENCE **35** (2000) 2443 – 2449

The cracking of zirconia refractory tubes under hot shock

PENGFEI HE
Department of Engineering Mechanics & Technology, Tongji University, Shanghai 200092, People's Republic of China

TIAN J. LU*
Cambridge University Engineering Department, Cambridge, CB2 1PZ, UK
E-mail: TJL21@eng.cam.ac.uk

WILLIAM J. CLEGG
Materials Science & Metallurgy Department, Cambridge University, Cambridge, CB2 3QZ, UK

The evolution of stresses and strains in a zirconia-containing refractory tube subjected to a hot shock on the outer surface and convective cooling at the inner surface is analysed with the method of finite elements. To account for the temperature-induced phase transformation in the zirconia as well as the overall thermal expansion, a coefficient of total dilatation is introduced. The parameters that control the time-dependent stress and strain responses are identified by performing finite element calculations that span the range of variables relevant to steel making. The effects of tube thickness, hot shock duration, initial temperature, temperature dependence of elastic modulus, and transformation amplitude on stress and strain distributions are discussed, and heating and process strategies to eliminate surface cracking are suggested.

1. Introduction

Zirconia (ZrO_2) is widely used in refractory applications (e.g., slag line sleeves for steel making) due to its high corrosion resistance, high refractory index, high hardness and low extinction coefficient. However, zirconia is unstable when subjected to thermal transients, which may cause cracking in the refractory [1]. At room temperature, bulk zirconia with large grain size exhibits typical monoclinic structure. Upon heating to about 1150°C, the monoclinic phase transforms to the tetragonal phase, which is stable up to about 2300°C, before it transforms to the cubic phase [2]. The above transformation process is reversed when zirconia is cooled from a high temperature (>2300°C) to room temperature.

Extensive cracking has been observed in the manufacturing of large AZS (alumina/zirconia/silica) blocks: here, cracks form under cooling from the casting temperature. A computational model, based on the Drucker-Prager plasticity theory, has been successfully used to study the fracturing behaviour of an AZS block subjected to surface cooling [1]. It is concluded that the cracking is attributable to the tetragonal-to-monoclinic phase transformation during cooling. Specifically, it is demonstrated that geometrical constraint from the centre on transformational dilatation at the surface, which commences when the temperature there drops below 1000°C, causes inelastic dilatation near the centre. These strains, which arise through cavitational damage, can exceed the rupture strain for the material, which is especially small at the higher temperatures existing in the centre. The cracks formed when the rupture strain has been exceeded have large opening displacements, because of the viscous flow occurring at these relatively high temperatures. The band of material experiencing tensile stress moves gradually outwards as the surface temperature drops below 1000°C, with the surface layer eventually experiencing hoop tension. However, these stresses only arise when the surface is quite cool (∼700°C) and there is no experimental evidence that cracking occurs at such temperatures, wherein the material is elastic and brittle. If the refractory block does not contain zirconia, then surface cracking rather than cracking near the centre is more likely to occur under rapid cooling (i.e., cold shock) [4].

Experimentally, it is found that the slag line sleeves made of zirconia-containing refractory suffer from extensive surface cracking upon sudden heating from room temperature up to about 1500°C (i.e., a hot shock). These surface cracks not only limit significantly the service life of a sleeve but also provide passage ways for the relatively rapid attacking of corrosive agents, and hence should be suppressed. In the absence of zirconia, a refractory block subjected to a hot shock develops tensile stress at the centre and compressive stresses near the surface, and hence surface cracking is, normally, unlikely to occur [4]. Therefore, analogous to

* Author to whom all correspondence should be addressed.

the case of an AZS block under cooling, it is believed that the observed surface cracking of a slag line sleeve made of zirconia-containing refractory subjected to a hot shock is closely linked to the transformation of zirconia from monoclinic phase to tetragonal phase. This paper presents a finite element study on the evolution of stresses and strains in a zirconia refractory tube subjected to a hot shock on the outer surface and convective cooling at the inner surface, mimicking the conditions experienced by a slag line sleeve during service. The effects of heat-up time, tube thickness, phase transformation amplitude, and temperature dependence of elastic modulus are discussed, and heating strategies to eliminate surface cracking are suggested. The zirconia refractory considered in the present work behaves linear elastically at temperatures up to 1500°C, in sharp contrast to the constitutive behaviour of the AZS material, suggesting the development of a computational model different from the Drucker-Prager model for AZS.

2. The model

Consider a long cylindrical tube made of zirconia refractory with inner radius R_i and outer radius R_o (Fig. 1). Let the tube has a uniform initial temperature T_0. At time $t = 0$, the outer surface of the tube is heated according to

$$T_\mathrm{s}(t) = T_0 + (T_\mathrm{G} - T_0)\frac{t}{t_\mathrm{G}} \qquad (1)$$

where T_s is the temperature of the outer surface, T_G is the goal temperature and t_G is the total ramp-up time that is needed to heat the outer surface from T_0 to T_G. A cylindrical coordinate system (r, θ, z) located at the centre of the tube is introduced, with the z-axis lying along the longitudinal direction. Without loss of generality, plane strain and axisymmetrical deformation of the tube is assumed such that the tube temperature may be denoted by $T(r, t)$. The conduction of heat in the tube is then governed by

$$\frac{\partial^2 T(r,t)}{\partial r^2} = \frac{1}{\kappa}\frac{\partial T}{\partial t} \qquad (2)$$

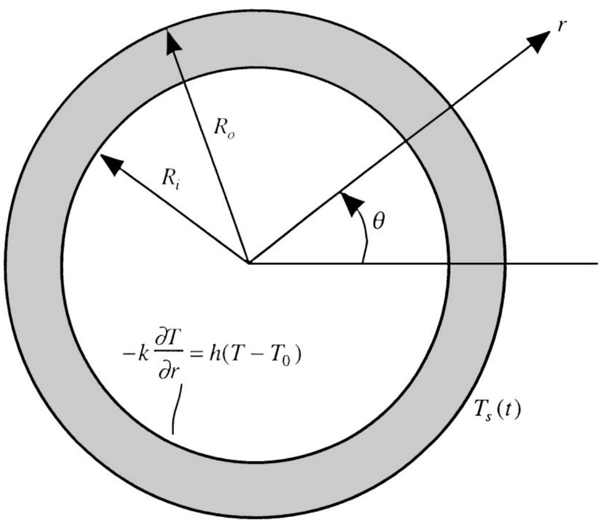

Figure 1 Section of the tube and boundary conditions.

where κ is the thermal diffusivity of the refractory. Both κ and the thermal conductivity k will be taken as temperature independent. The inner surface of the tube is subjected to (natural) convective cooling, with

$$-k\frac{\partial T(r,t)}{\partial r} = h(T(r,t) - T_0), \quad \text{at} \quad r = R_\mathrm{i} \qquad (3)$$

The initial condition of the problem is

$$T(r, 0) = T_0 \qquad (4)$$

The zirconia-containing refractory is assumed to be isotropic and linearly elastic up to 1500°C, as evidenced by available experimental data. Let E, α, c, ρ, ν denote the Young's modulus, coefficient of thermal expansion (CTE), specific heat (at constant pressure), density, and Poisson's ratio for the refractory, respectively. The dependence of E and α on temperature will be accounted for, thus $E = E(T)$ and $\alpha = \alpha(T)$. The stresses and strains developed in the tube due to thermal transients are denoted, in the cylindrical coordinates, as $(\sigma_r, \sigma_\theta, \sigma_z)$ and $(\varepsilon_r, \varepsilon_\theta, \varepsilon_z)$. At temperatures around 1160–1200°C, the phase transformation of the monoclinic zirconia to tetragonal takes place in the refractory. (Zirconia may also undergo m-t or t-m transformation under pressure or shear forces, or both. The stress-induced phase transformation is neglected in the present work.) The dilatational transformation strain arising from the phase transformation is denoted here by ε^T, which depends on the temperature T and on the volume fraction of the transforming phase at radial position r:

$$\varepsilon^T(r, T) = g(T)\varepsilon_0^T(r)$$

with $g(T)$ denoting the fraction of the phase transformed at T and $\varepsilon_0^T(r)$ the maximum dilatational transformation strain that can be achieved at r. In accordance with ABAQUS (a commercially available finite element code) specifications, the model employs the total "transformation strain" as the sum of the transformation and the thermal strains. With the initial processing temperature, T_0, chosen as the reference from which the strains are measured, the total strain is

$$\varepsilon_\mathrm{total}^T(r, T) = \varepsilon^T(r, T) + \int_{T_0}^{T} \alpha(\tilde{T})\, d\tilde{T} \qquad (5)$$

To comply with ABAQUS, a secant CTE is defined as:

$$\alpha_\mathrm{secant}(T) = \varepsilon_\mathrm{total}^T(T)/(T - T_0) \qquad (6)$$

such that, at given time t and radial position r, the strain increments are given by

$$\dot{\varepsilon}_{ij} = \frac{1+\nu}{E}\dot{\sigma}_{ij} - \frac{\nu}{E}\dot{\sigma}_{kk}\delta_{ij} + \alpha_\mathrm{secant}\dot{T}\delta_{ij} \qquad (7)$$

Here, $i = r, \theta, z$, the summation convention over repeated indices applies, and δ_{ij} is the Kronecker delta.

The experimentally measured $\varepsilon_\mathrm{total}^T(T)$ versus temperature T curve is shown in Fig. 2a for two selected

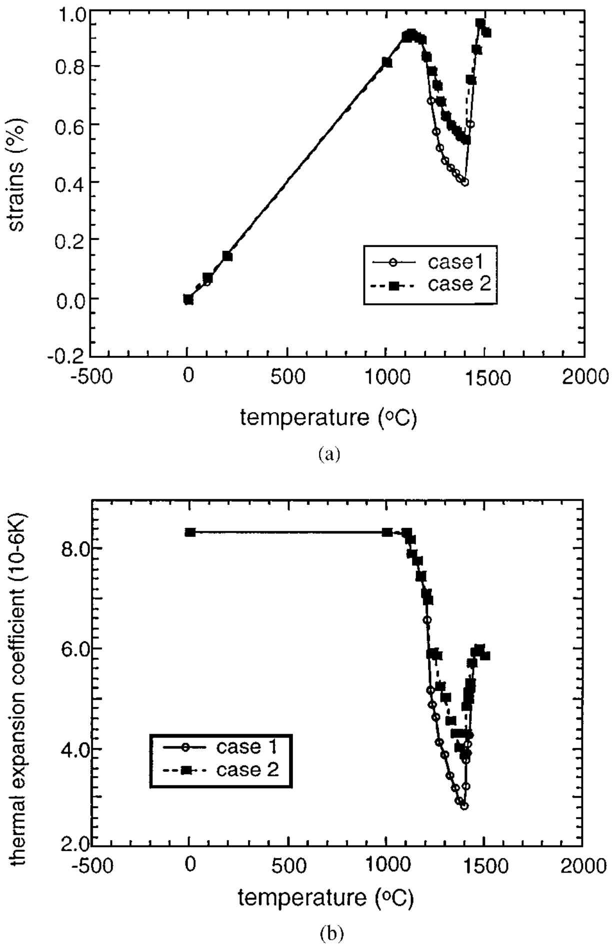

Figure 2 (a) Total dilatational strain, (b) secant coefficient of thermal expansion, as functions of temperature for both cases 1 and 2.

cases: case 1 with $\varepsilon_0^T = 0.5\%$ and case 2 with $\varepsilon_0^T = 0.35\%$, both representative of slag line sleeves. The variation of ε_0^T with r is small, and hence is neglected in the present investigation. The corresponding α_{secant} versus T curve is plotted in Fig. 2b. Note that, in both cases, the material has a relatively constant CTE of $\sim 8.5 \times 10^{-6}\ \text{K}^{-1}$ up to about 1100°C. The monoclinic-to-tetragonal phase transformation starts at 1160°C and finishes at about 1200°C. The secant CTE drops to a minium at about 1400°C, reaching $\sim 3 \times 10^{-6}\ \text{K}^{-1}$ in case 1 and $\sim 4 \times 10^{-6}\ \text{K}^{-1}$ in case 2.

3. Numerical results

Under plane strain, there are no gradients of temperature, stresses and strains in the axial direction z. The problem is therefore modelled with a single row of 8-node quadratic axisymmetric elements both for the heat transfer and stress/strain analysis. A finer mesh is used towards the outer surface of the tube, where the transients of stresses and temperature are largest. The convective heat transfer boundary condition at the inner surface is simulated by the film technique provided in ABAQUS. To save computational time, the heat transfer problem is decoupled from the stress/strain problem: the transient temperature distribution from the heat transfer analysis is written to the results file, the latter being used as input to the subsequent stress/strain analysis. It has been established that the results obtained from the decoupled analysis agree excellently well with those calculated from a fully coupled temperature-displacement analysis.

For plane strain, $\varepsilon_z = 0$. Since the tube thickness of slag line sleeve is usually thin, σ_r is very small compared with other stress components. Hence, the focus below will be on the evolution of radial and hoop strains ε_r, ε_θ, and axial and hoop stresses σ_z, σ_θ.

3.1. Reference case

The problem corresponding to case 1 of Fig. 2 is computed first to provide baseline solutions, with the parameters selected as follows:

Thermal conductivity, k	$8\ \text{W/(m} \cdot \text{K)}$
Specific heat, c	$500\ \text{J/(kg} \cdot \text{K)}$
Density, ρ	$5800\ \text{kg/m}^3$
Inner radius of tube, R_i	0.05 m
Thickness of tube, H	0.005 m
Total ramp-up time, t_G	100 s (1.667 minute)
Goal temperature, T_G	1500°C
Initial temperature, T_0	20°C
Poisson's ratio, ν	0.2
Young's modulus, $E(T)$	100–130 GPa
Heat transfer coefficient, h	$10\ \text{W/m}^2\ \text{K}$

Without loss of generality, E is assumed to decrease linearly with increasing temperature T, with $E(T_0) = 130$ GPa and $E(T_\text{G}) = 100$ GPa. The above parameters are typical for a zirconia-containing refractory used in steel making.

The evolution of temperature with heating time t at the element immediately below the outer surface $r = R_\text{o}$ is plotted in Fig. 3a, which is seen to follow closely that given by Equation 1. The distribution of temperature across the tube thickness at $t = 81.5$ s (1.33 minute) is shown in Fig. 3b—the temperature of the inner surface is seen to be about 70°C lower than that of the outer surface. Plots of thermal stresses and strains against time t are presented in Fig. 4 for the surface element. As was discussed before, the monoclinic zirconia transforms to tetragonal phase at about $T = 1150$°C, corresponding to $t = 1.33$ minute in the present case. Before the transformation takes place, the hoop stress σ_θ is compressive but quite small, nearly independent of t, whereas the axial stress σ_z, also compressive, increases linearly with t. On the other hand, the radial strain ε_r is roughly the same as the hoop strain ε_θ, both increasing linearly with t. When the phase transformation of zirconia occurs, it brings about significant changes in the response behaviours of the tube. In particular, it is noted from Fig. 4a that the hoop stress σ_θ changes from compressive to tensile, with a maximum tensile hoop stress of about 225 MPa. This stress is expected to surpass the tensile failure stress of the zirconia-containing refractory ($\sim$110 MPa at room temperature). Consequently, cracks, extending in the axial direction perpendicular to σ_θ, may initiate from the outer surface under such severe tensile stressing.

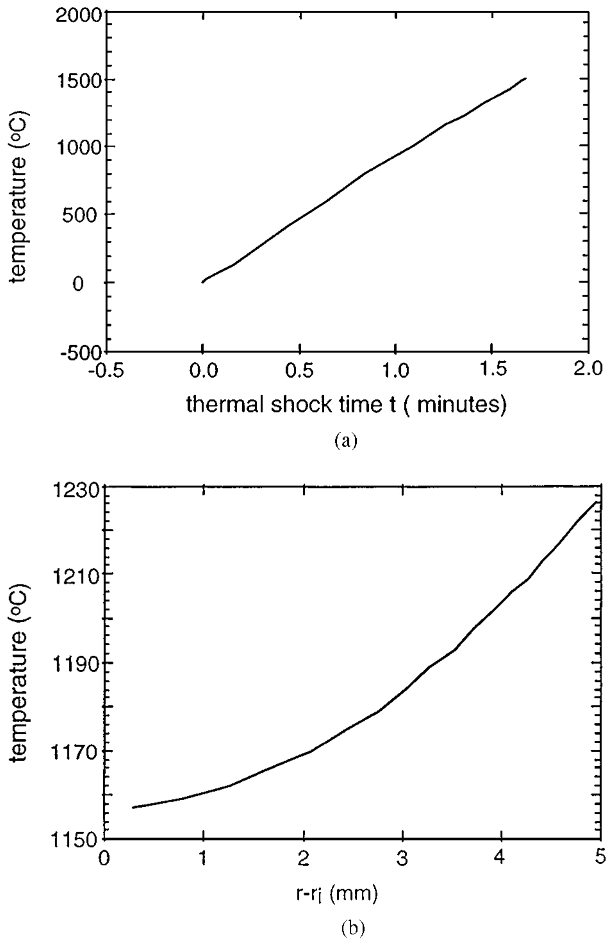

Figure 3 (a) Evolution of surface temperature, (b) temperature profile in the tube at time $t = 1.33$ minutes for case 1.

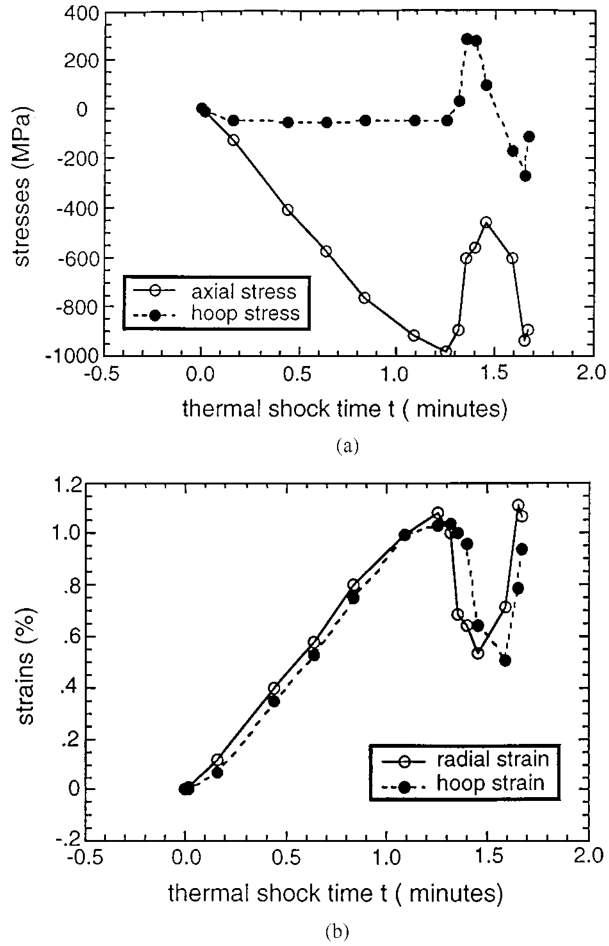

Figure 4 Evolution of (a) surface stresses, (b) surface strains for case 1.

The distribution of stresses and strains across the tube thickness at $t = 1.33$ minute, is shown in Fig. 5. The hoop strain is constant across the whole cylinder at this instant; the radial strain and the axial and hoop stresses are also constant except for the region close to the outer surface. Here, the hoop stress changes sign (from compressive to tensile) due to transformation effects. If the heating of the tube stops at $t = 1.66$ minute, only the surface layer will have experienced tensile stressing as the rest of the tube has yet to reach the transformation temperature. Consequently, cracks, once formed at the outer surface, is unlikely to extend into the interior of the tube because of the large compressive stresses there. However, if $t_G > 1.33$ minute, the band of material experiencing tensile stressing moves gradually from the outer surface towards the inner surface, causing the surface cracking to extend in the thickness direction.

3.2. Effect of tube thickness (H)

The influence of tube thickness H on thermal stress and strain distributions inside the tube is studied, with H varied in the range of 1 to 8 mm. Plots of stresses and strains near the outer surface as functions of time t are given in Fig. 6a and b for $H = 1$ mm and 5 mm, respectively. The rest of the parameters used for plotting Fig. 6 are identical to those for Fig. 4. The tube thickness H affects significantly the hoop stress σ_θ, which is more clearly seen from Fig. 7 where the maximum hoop stress $\sigma_\theta^{\max}$ is plotted as a function of H. Here, for a tube of given thickness H, $\sigma_\theta^{\max}$ is defined as the maximum hoop stress in the tube over all time t and position r. Fig. 7 reveals that the magnitude of the maximum hoop stress $\sigma_\theta^{\max}$ increases sharply as H is increased to about 4 mm, after which its rate of increase slows down. If $\sigma_\theta^{\max}$ is confined not to exceed 110 MPa, the tensile strength of the refractory, then the tube thickness must not exceed 2 mm (other parameters remain unchanged) in order to eliminate surface cracking.

3.3. Effect of total heating time (t_G) and initial temperature (T_0)

With the goal temperature fixed at $T_G = 1500°C$ and tube thickness fixed at $H = 5$ mm, the effect of changing the total heating time t_G on $\sigma_\theta^{\max}$ is displayed in Fig. 8, with the initial temperature T_0 fixed at $T_0 = 20°C$; the remaining parameters used for the plotting are the same as those for Fig. 4. As expected, $\sigma_\theta^{\max}$ decreases considerably as the total heat-up time is increased from 1 minute to 20 minutes, although the knock-down in $\sigma_\theta^{\max}$ is not sufficient to prevent cracking in this particular case. Also, we have found that pre-heating the tube to $T_0 = 1100°C$ before a hot shock is applied has little effect in reducing $\sigma_\theta^{\max}$, if the total heating time is less than about 20 minutes. Consequently, additional

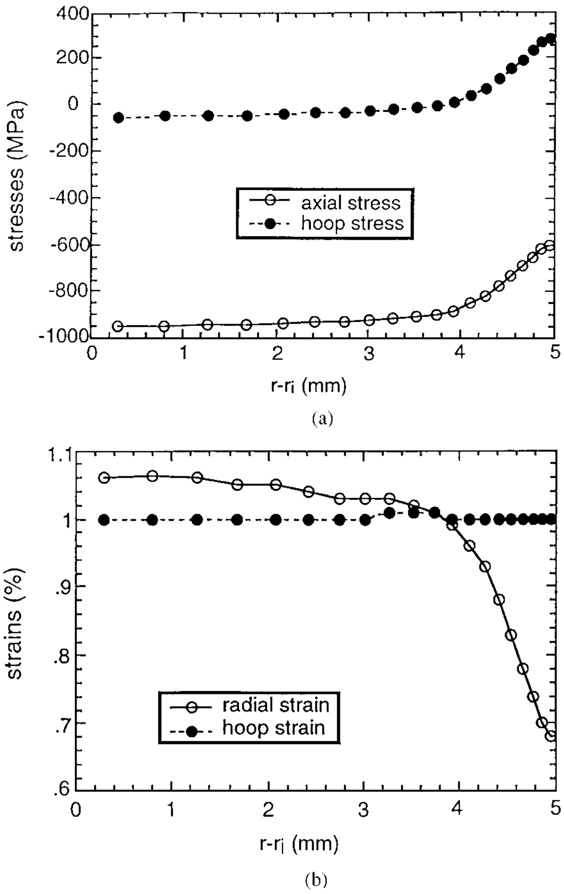

Figure 5 Variation of (a) stresses, (b) strains across tube thickness at time $t = 1.33$ minute for case 1.

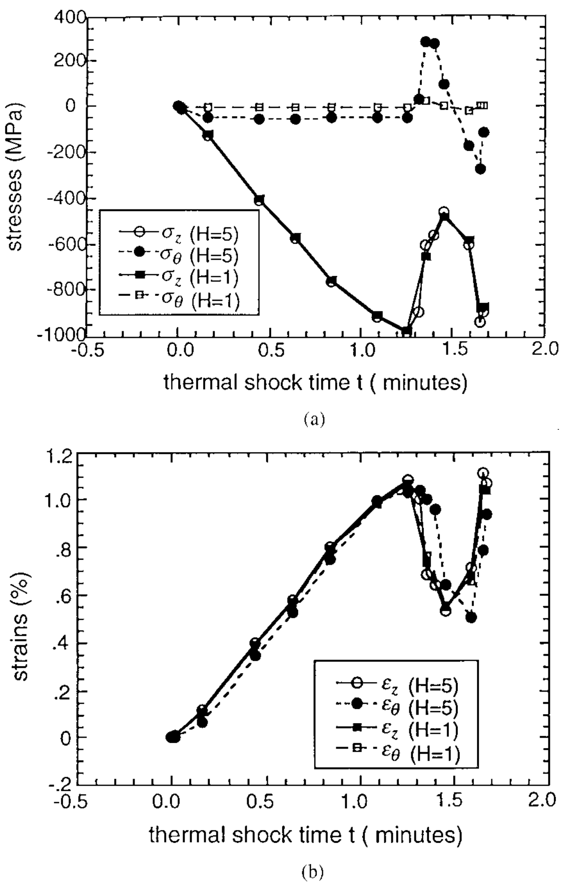

Figure 6 Effect of tube thickness H on evolution of (a) surface stresses, (b) surface strains.

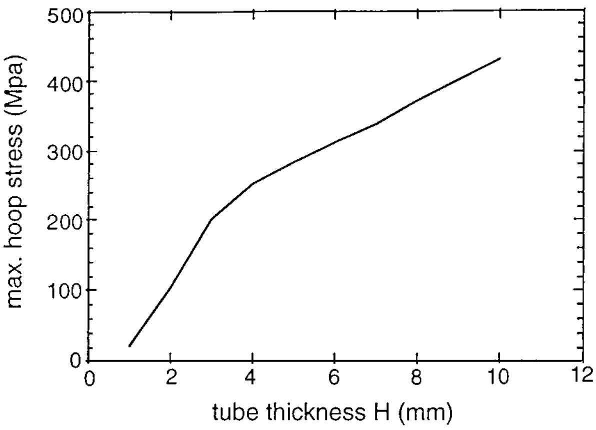

Figure 7 Effect of tube thickness H on maximum hoop stress.

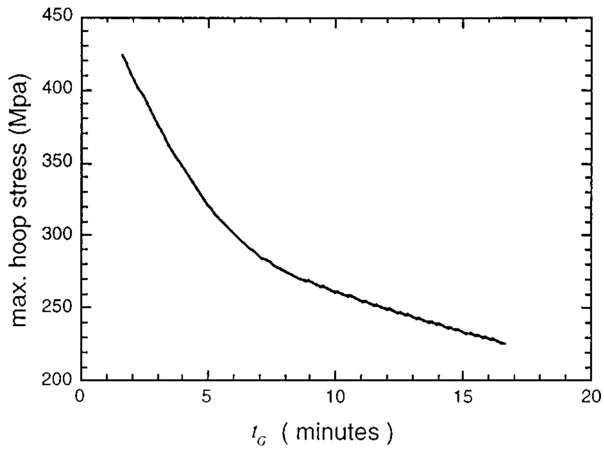

Figure 8 Effect of initial temperature T_0 and total heating time t_G on maximum hoop stress.

strategies such as reducing the tube thickness must be introduced if cracking is to be suppressed.

3.4. Effect of phase transformation amplitude (ε_0^T)

The effect of phase transformation amplitude ε^T on thermal transient stresses and strains is studied for three cases: $\varepsilon_0^T = 0.5\%$ and $\varepsilon_0^T = 0.35\%$ (cases 1 and 2 in Fig. 1) and $\varepsilon_0^T = 0$ (case 3). The results for material elements adjacent to the outer surface are shown in Fig. 9. As expected, all the changes in the stress and strain distributions can be traced back to phase transformation in zirconia, with case 1 providing the largest tensile hoop stress. In the case of zero transformation (case 3), all the stress components in material elements near the outer surface are compressive during the whole course of heating, and hence will not cause surface cracking—however, as previously mentioned, cracking may occur in the interior part of the tube due to the build-up of tensile stresses there under a hot shock [3].

3.5. Effect of $E(T)$

Finally, the effect of the Young's modulus varying with temperature on stress/strain evolution is studied. Fig. 10

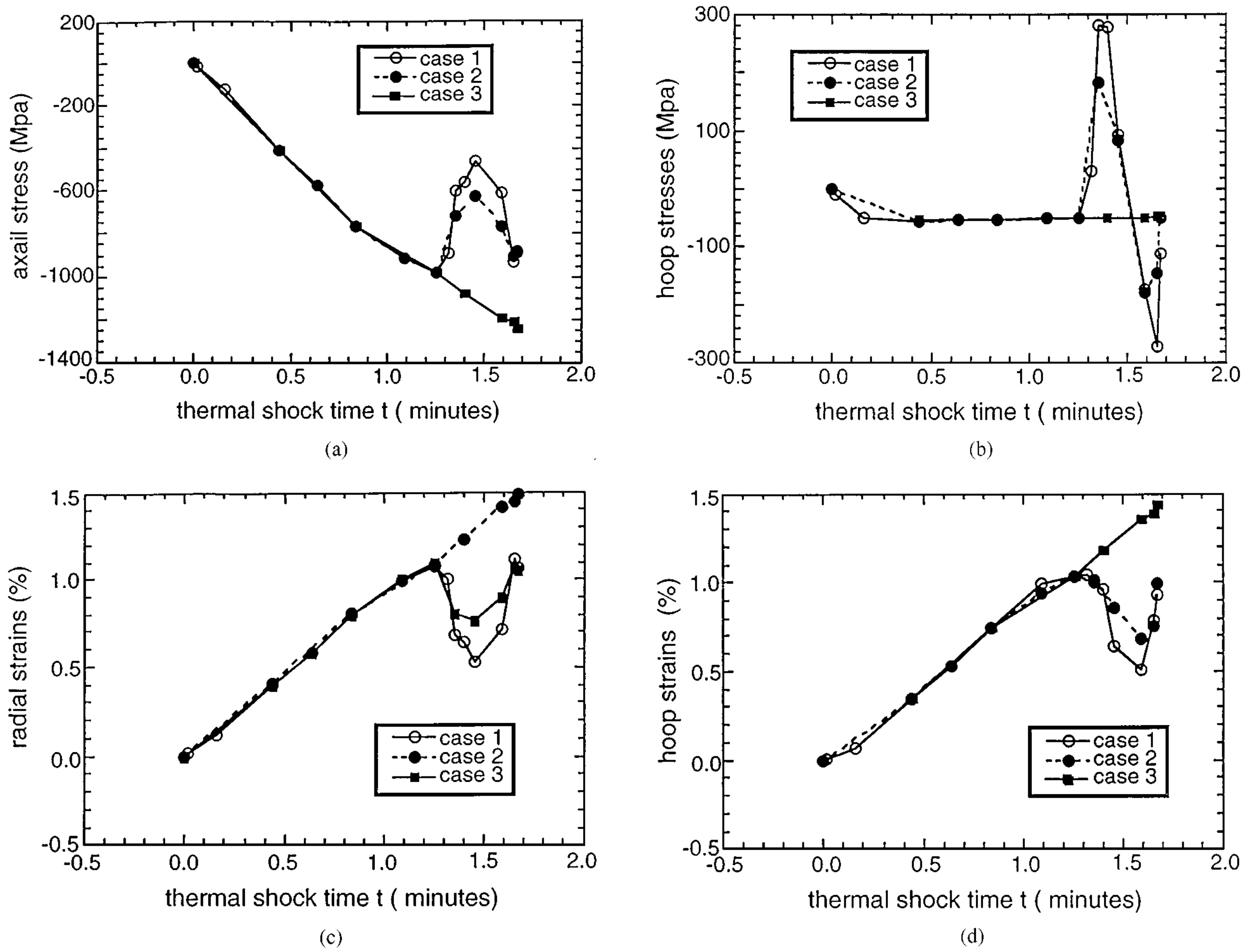

Figure 9 Effect of phase transformation amplitude ε^T on evolution of (a) axial stress, (b) hoop stress, (c) radial strain, (d) hoop strain.

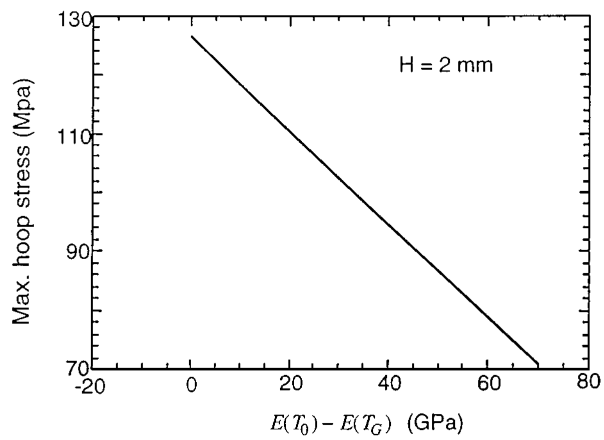

Figure 10 Effect of Young's modulus $E(T)$ on maximum hoop stress for case 1 with $H = 2$ mm.

presents the maximum hoop stress $\sigma_\theta^{\max}$ for a tube of thickness $H = 2$ mm as a function of $E(T_0) - E(T_G)$, the loss of the Young's modulus due to heating. For the plotting, the CTE corresponding to case 1 of Fig. 2b is used, the rest of the parameters being those listed for Fig. 4. It is seen from Fig. 10 that the maximum hoop stress $\sigma_\theta^{\max}$ decreases linearly as $E(T_0) - E(T_G)$ increases. For a 2 mm thick tube with a tensile strength of 116 MPa, surface cracking will occur if there is no degradation of the Young's modulus upon heating (i.e., $E(T_0) = E(T_G)$), but cracking will be suppressed if $E(T_0) - E(T_G)$ drops below 20 GPa.

4. Concluding remarks

If the outer surface of a tube made of zirconia-containing refractory is suddenly heated from room temperature to about 1500°C whilst its inner surface is cooled by natural convection, surface cracking is likely to occur in a tube due to the transformation of zirconia from monoclinic phase to tetragonal phase at about 1150°C. Such cracking happens when the maximum hoop stress near the outer surface of the tube exceeds the tensile strength of the refractory, and may extend gradually towards the inner surface if the total heat-up time is sufficiently long. A variety of approaches may be employed to eliminate the cracking, the most efficient being to reduce the tube thickness H, followed in ranking by prolonging the total time of heating t_G and reducing the phase transformation amplitude ε_0^T; pre-heating the tube to a temperature just below the transformation temperature (∼1150°C) alone does not appear to help the prevention of cracking, but the drop in Young's modulus as the refractory is heated potentially may have a significant knock-down effect on the maximum hoop stress that can be achieved in the tube.

The main outcome of the present study is perhaps the finding that cracking would be difficult, if not impossible, to prevent if a fully or partially stablizd ZrO_2 refractory tube is subjected to severe hot shock, which has been a long standing problem in steel making. This creates a dilemma, as the crack-inducing ZrO_2 (due to its phase transformation) is needed for its excellent corrosion resistance. Fortunately, one novel approach, developed recently at Cambridge, has shown considerable promise—the slag line sleeve now is made of ZrO_2-containing ceramic laminate with crack deflecting, porous interlayers, and has been demonstrated to have superior thermal shock as well as corrosion resistance. The key is that each lamina in the ceramic laminate is thin, with thickness typically less than 100 μm.

Acknowledgement

The stay of PH at Cambridge University was supported by an EC project.

References

1. T. J. LU, A. G. EVANS, J. W. HUTCHINSON, G. V. SRINIVASAN and S. WINDER, *J. Am. Ceram. Soc.* **81** (4) (1998) 917.
2. P.-Y. CHU, I. CAMPION and R. C. BUCHANAN, *J. Mater. Res.* **7**(11) (1992) 3065.
3. T. J. LU and N. A. FLECK, *Acta Mater.* **46** (1998) 4755.
4. L. G. ZHAO, T. J. LU and N. A. FLECK, *J. Mech. Phys. Solids*, in press.

Received 27 May
and accepted 23 November 1999