ON TRANSIENT IRRADIATION BEHAVIOR OF HTGR FUEL PARTICLES

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SUMMARY

An examination of HTGR TRISO coated fuel particles was made in which the particles' stress-strain histories were determined during both steady-state and transient operating conditions. The basis for the examination was a modified version of a computer code written by Kaae which assumed spherical symmetry, isotropic thermal expansion, isotropic elastic constants, time-temperature-irradiation invariant materials properties, and steady state operation during particle exposure. Additionally, the Kaae code modelled potential separation of layers at the SiC-inner PyC interface and considered that several entrapped fission products could exist in either the gaseous or solid state, dependent upon particle operating conditions.

The Kaae code was modified to study the effects of each of the following in the prediction of layer stresses:

(1) changes in Young's modulus of elasticity for both PyC and SiC with fluence and/or temperature;
(2) temperature dependence of the internal fission gas pressure calculations;
(3) a second-order correction to the residual thermal stress calculation;
(4) sensitivities of the stresses to phenomena modelled.

Using the modified code which modelled transient behavior in a quasi-static fashion, a series of both steady-state and transient operating condition computer simulations was made. For the former set of runs, a candidate set of particle dimensions and a nominal set of materials' properties was assumed. Layer thicknesses were assumed to be normally distributed about the nominal thicknesses and a probability distribution of SiC tensile stresses was generated; sensitivity of the stress distribution to assumed standard deviation of the layer thicknesses was acute. Further, this series of steady-state runs demonstrated that for certain combinations of the assumed PyC-SiC bond interface strength and irradiation-induced creep constant, anomalous predicted stresses may be obtained in the PyC layers. The steady-state runs also suggest that transient behavior would most likely not be significant at fast neutron exposures below about 10^{21} NVT due to both low fission gas pressure and likely beneficial interface separation.

The transient series of simulations assumed a 3 °C/sec rate of temperature increase lasting 200 seconds placed at various points in time; both constant and fluence/temperature varying materials properties were considered. The following set of conclusions were drawn based upon these runs.

(1) Only a minor increase in failure at a given burnup is expected above that fraction predicted by steady-state runs.
(2) Inclusion of environment-dependent materials properties yields, at best, a second order correction to constant-property results.
(3) Early-life predicted layer stresses are strongly dependent upon buffer and kernel thicknesses, creep constant, and SiC-PyC interface bond strength.
(4) Late-life predicted stresses are most strongly dependent upon buffer and kernel thicknesses and creep constant; physically realistic interface bond strengths do not greatly affect late-life predicted stresses.
(5) Volatilization of condensed fission products at elevated temperatures may be significant in fission gas pressure calculations.

1. Introduction

A computer code [1] was modified to study the stress-strain-irradiation histories of TRISO HTGR multi-layer fuel particles. Since the code modification, to a large extent, did not involve major changes to either the particle physics or the simplifying assumptions inherent in the original computer model, these basic features will be briefly stated for the purpose of completeness.

1. Spherical symmetry is assumed.
2. Elastic properties and thermal expansion are assumed to be isotropic.
3. The innermost, porous buffer layer does not possess mechanical strength; internal fission gas pressure is assumed to act at the inner edge of the innermost dense pyrocarbon (PyC) layer.
4. Silicon carbide (SiC) is assumed to be dimensionally stable under fast-neutron irradiation; PyC is assumed to have anisotropic irradiation-induced dimensional changes as described by input data.
5. Certain fission products may exist in either a gaseous or a condensed state, determined by the temperature-dependent allowable partial pressure for the species.
6. Coating layers may transmit stresses to each other due to applied internal and/or external pressures, by temperature expansion/contraction displacement differences, and due to irradiation-induced dimensional changes.
7. Stresses may be relieved by irradiation-induced creep. Thermally induced creep is ignored as it is significantly smaller than irradiation-induced creep.

The unmodified code did not, however, allow material properties to vary with temperature and/or irradiation and did not permit simulation of particle irradiation histories during which temperature and/or neutron flux varied with time. The code was modified specifically to allow such histories to be simulated with the specific intent of predicting stresses during both steady state and transient operation conditions [2].

2. The Particle Model

Combining the equation of equilibrium with elastic stress-strain relations, and treating the inelastic strains due to temperature and irradiation as forcing functions, one obtains the basic differential equation for the radial displacement, U(r), as

$$r^2 \frac{d^2U(r)}{dr^2} + 2r\, U(r) - 2\, U(r) = P(r) \qquad (1)$$

where

$$P(r) = \frac{2r(2\mu - 1)[G_T(r) - G_r(r)]}{1 - \mu} + r^2\left[\frac{dG_r(r)}{dr} + \frac{2\mu}{1 - \mu}\,\frac{dG_T(r)}{dr}\right] \qquad (1a)$$

and where

$G_T(r)$ = Tangential strain due to thermal and irradiation effects

$G_r(r)$ = Radial strain due to thermal and irradiation effects

This non-homogenous Euler (or non-dimensional) equation may be solved by any number of means, but the authors used the method of variation of parameters to obtain

$$U(r) = \frac{r}{3}\int_a^r \frac{P(r)}{r^2}\,dr - \frac{r^{-2}}{3}\int_a^r r\,P(r)\,dr + C_1 r + C_2 r^{-2} \qquad (2)$$

One determines the values of C_1 and C_2 from the appropriate boundary conditions;

specifically, for a spherical shell with inner radius r_i and outer radius r_o, and applied pressures of P_1 and P_o, respectively, the boundary conditions would be

$$\sigma_r(r_i) = -P_1 \qquad \text{and} \qquad \sigma_r(r_o) = -P_o \tag{3}$$

and the inner and outer displacements, $U(r_i)$ and $U(r_o)$, respectively, are given by

$$U(r_i) = \frac{r_i r_o^3}{r_o^3 - r_i^3} \int_{r_i}^{r_o} \frac{[G_T(r) - G_r(r)]}{r}\, dr + \frac{r_i}{r_o^3 - r_i^3} \int_{r_i}^{r_o} [2\, G_T(r) + G_r(r)] r^2\, dr + \frac{(2 - 4\mu)(P_2 r_i^{-2} - P_1 r_o^{-3} r_i) + (1 + \mu) r_i^{-2} (P_2 - P_1)}{2E(r_o^{-3} - r_i^{-3})} \tag{4}$$

$$U(r_o) = \frac{r_o r_i^3}{r_o^3 - r_i^3} \int_{r_i}^{r_o} \frac{[G_T(r) - G_r(r)]}{r}\, dr + \frac{r_o}{r_o^3 - r_i^3} \int_{r_i}^{r_o} [2\, G_T(r) + G_r(r)] r^2\, dr + \frac{(2 - 4\mu)(P_2 r_i^{-3} r_o - P_1 r_o^{-2}) + (1 + \mu) r_o^{-2} (P_2 - P_1)}{2E(r_o^{-3} - r_i^{-3})} \tag{5}$$

where

μ = Poisson's ratio for the material in the shell

E = Young's modulus for the material in the shell

For the multi-layer TRISO particles, with the simplifying assumptions made, there are four boundaries at which conditions are imposed:

) PyC) SiC) PyC)

r_1 r_2 r_3 r_4

a. $\sigma(r_1)$ = - Fission gas pressure

b. $\sigma(r_4) = 0$

c. $\sigma(r_3)$ = Pressure due to displacement mismatch of SiC and outer PyC

d. $\sigma(r_2)$ = Pressure due to displacement mismatch of Sic and inner PyC <u>or</u> $\sigma = 0$ if interface separation occurs

These displacement terms are all calculable and combination of the displacements with the stress-strain relations and strain displacement relations yields the stress equations of Reference [1]. However, a modification is made by the authors in the calculation of residual thermal stress. From the isotropic thermal expansion

$$G_T(r) = G_R(r) = \alpha \Delta T$$

we obtain

$$\frac{1}{r_o^3 - r_i^3} \int_{r_i}^{r_o} [2\, G_T(r) + G_r(r)] r^2\, dr = \alpha \Delta T$$

$$\therefore\ U(r_i) = \alpha r_i \Delta T \qquad \text{and} \qquad U(r_o) = \alpha r_o \Delta T$$

Assuming that SiC is deposited upon the inner PyC at temperature T_{C1} and that each coating has a [formal] reference temperature, then there is an interface displacement mismatch at an arbitrary temperature of

$$\delta U(r_2) = r_2(\alpha_2 - \alpha_1)(T - T_{C2}) \tag{6}$$

Addition of another layer deposited at $T = T_{C3}$ has no external applied stresses, but an internal residual thermal stress calculable from the temperature differences. This residual stress, P*, is given by

$$P^* = \frac{(\alpha_2 - \alpha_1)(T_{C3} - T_{C2})}{\dfrac{(4\mu_1 - 2)r_2^3 - (1 + \mu_1)r_1^3}{2E_1(r_2^3 - r_1^3)} + \dfrac{(4\mu_2 - 2)r_2^3 - (1 + \mu_2)r_3^3}{2E_2(r_3^3 - r_2^3)}} \tag{7}$$

from which the mismatch at r_3 is given by

$$\delta U(r_3) = r_3(T - T_{C3})(\alpha_3 - \alpha_2) - \frac{3}{2}\,\frac{P^*(1 - \mu_2)r_3 r_2^3}{E_2(r_3^3 - r_2^3)} \tag{8}$$

It may be noted that this differs from the expression given in Reference [1] only by the last term and, further, that the correction term is zero if either the coefficients of expansion or deposition are equal.

Materials properties were assumed to vary with temperature and fluence in the following fashion. Young's modulus of elasticity for PyC was assumed to increase linearly with respect to fast neutron irradiation from an unirradiated value of 4.0×10^6 psi to 8.0×10^6 psi at nvt = 4×10^{21}, and remained constant for higher exposures. Young's modulus of elasticity for SiC was assumed to decrease linearly with temperature from a room temperature value of 6.0×10^7 psi to a value of 3.0×10^7 psi at a temperature of 1,500° C.

With these model changes and with the changes incorporated to allow the specification of temperature and flux time histories to be input, the code was used to predict stress histories in a quasi-static fashion (i.e., assuming average temperature and flux levels over a series of time increments). A series of both steady-state and transient computer simulations was made; the former were made to explore the sensitivity of the stresses to various phenomena, the latter to determine over what regimes these sensitivities might change for transient analyses.

3. Results of the Computer Simulations

For the steady-state computer runs, a nominal set of particle dimensions [3] was assumed and a fixed set of anisotropic irradiation-induced dimensional change data was employed; the appropriateness of these data to match all operating conditions was assumed although such an assumption is clearly an approximation. An initially large (8×10^4 psi) value for the inner PyC-SiC interface bond-strength was employed and sensitivity to the creep constant was examined. For those runs employing low values of the creep constant, tensile stresses were developed in the PyC which were unquestionably higher than the tensile fracture stress of PyC, and the predicted stresses do not, therefore, stand up to close scrutiny. It seems reasonable (as well as practical) to limit the bond strength of the interface to, at most, the tensile stress of PyC for simulations in which the creep constant is insufficiently large to relieve stresses before they become prohibitively large.

Assuming that particle failure would be related to particles having high tensile stress in their SiC layers, a series of runs was made in which particle buffer and kernel thicknesses were assumed to vary from their nominal values and were irradiated to high burnup. The very early time (burnup) behavior appeared to be relatively insensitive to the assumed creep constant, as one might expect from the assumed form of the creep law. Interface bond strength becomes important shortly thereafter, since weak bonds will quickly allow separation as the PyC shrinks away from the SiC. At late times the bond strength becomes a relatively unimportant parameter as the SiC-PyC interface recloses as internal pressures increases.

A typical end-of-life peak SiC tensile stress distribution for various assumptions regarding the fractional standard deviation in buffer and kernel dimensions is given in Figure 1. It seems clear that prediction of the failed fraction of the fuel at the end of life is strongly dependent upon how well one can quantify the distribution of particle dimensions.

Computer simulations of transients all assumed a 3° C/sec rate of temperature increase lasting for 200 seconds with the reactor initially at 900° C. Placement of the transient early in the fuel particle lifetime did not cause SiC tensile stresses of the order of 3×10^4 psi since insufficient fission gas was entrapped to cause a large increase in the inner boundary stress value. However, for placements of the transient late in the particle lifetime an increase in SiC tensile stress above the former end-of-life stress was noted. Such an increase is very dependent upon the assumed magnitude of the transient and the operating condition at (as well as the irradiation history up to) the time of the transient. At this time insufficient data for a class of transients and operation histories has been generated to enable a redrawn version of Figure 1.

The temperature transients pointed out two features of the models employed. First, the inclusion of condensable fission products tends to increase the expected value of the inner pressure because at elevated temperatures the allowable partial pressures for the fission products Cs, Rb and Te allow previously condensed "globs" of these substances to become gaseous and, although this increases the volume available to accomodate fission gas, the net increase in the number of moles of fission gas accounts for a net increase in the pressure. The magnitude of this difference is indicated in Figure 2.

Further, inclusion of environment-dependent materials properties as described previously may or may not affect predicted stresses during a transient. For example, Figure 3 depicts differences predicted by allowing different values of Young's modulus for both SiC and PyC than our assumed model. In truth, the "nominal model" curve also contains data from two other simulations which essentially plot into the same curve. It appears that our model becomes most significant when it most strongly alters the relative magnitude of the ratio of E_{SiC} to E_{PyC}; environmental conditions from which weak variations in this ratio arise will not significantly alter predicted stresses. It is also worthy of mention that although Figure 3 clearly shows a difference, the relative difference of the models is small; a preliminary judgment might be that except for some as yet undetermined pathological examples, reasonably slight variations in materials properties do not greatly affect predicted stresses.

References

[1] KAAE, J. L., "A Mathematical Model for Calculating Stresses in a Pyrocarbon- and Silicon Carbide-Coated Fuel Particle," J. of Nucl. Materials, 29, pp. 249-266 (1969).

[2] MORTENSON, S. C., "Coated Particle HTGR Fuel Modeling and Its Application to Transient Analyses," Master's thesis in Engineering, University of California, Los Angeles (September 1976).

[3] SMITH, C. L., "Fuel Particle Behavior under Normal and Transient Conditions," Gulf Atomic Energy Company, San Diego, California, GA-A-12971 (October 1, 1974).

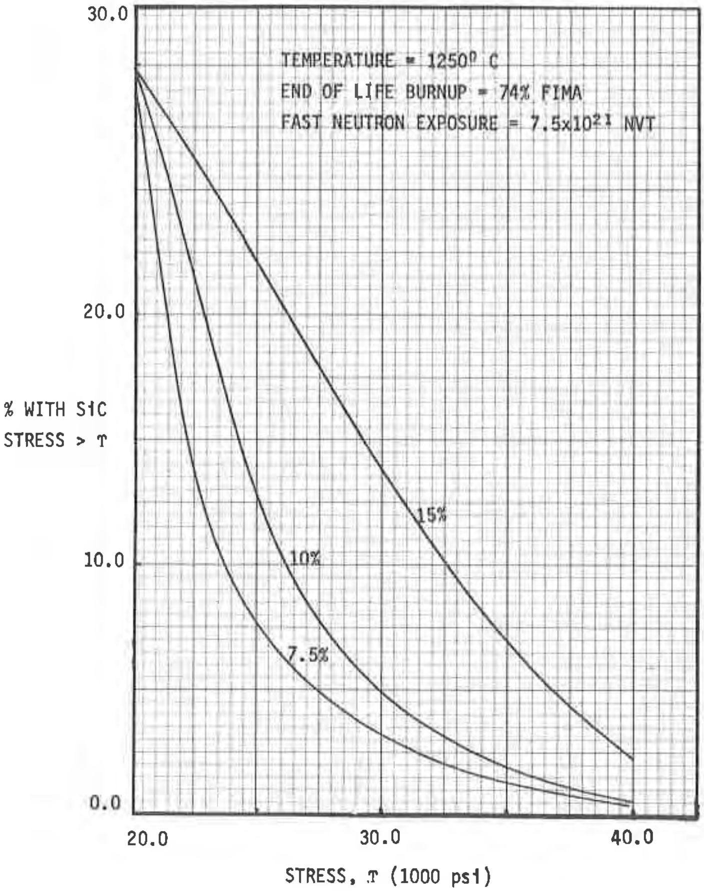

Figure 1. Sensitivity to Assumed Standard Deviation in Buffer and Kernel.

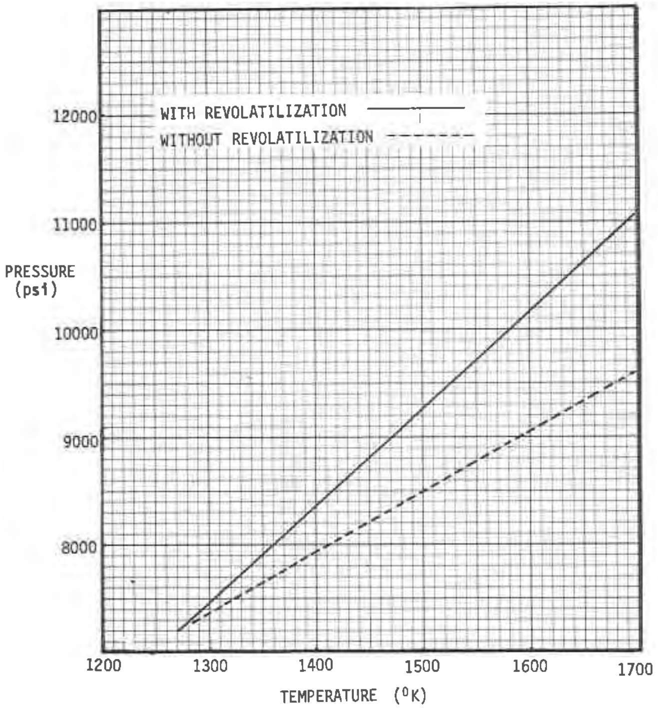

Figure 2. Typical Late Life Fission Gas Pressure During a Transient Temperature Increase Both with and without the Revolatilization of Condensables.

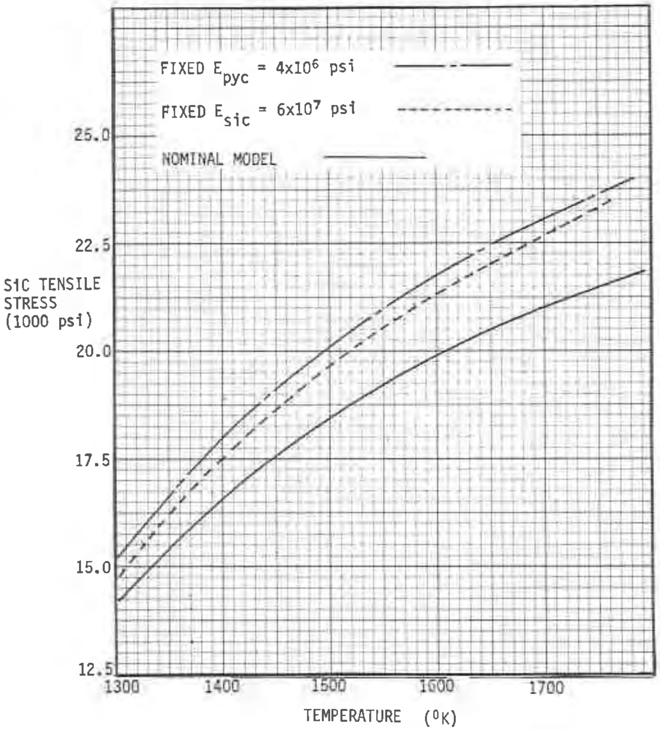

Figure 3. Variation of SiC Tensile Stress during a Temperature Transient for Various Materials Properties.