

MODELING OF LMFBR FUEL CLADDING MECHANICAL BEHAVIOR UNDER TRANSIENT CONDITIONS

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SUMMARY

Analytic methods must address the complexity of modeling fuel cladding response in situations where temperature, temperature gradient, strain rate, and stress are changing rapidly with time. Fuel cladding modeling is not a simple extension of the experimental and interpretative methods used in classical mechanics. New concepts are needed to describe materials in a reactor environment. Some aspects of continuum physics used to develop these new constitutive equations for fuel cladding are presented in this report.

The theory of internal state variable, whose evolution is governed by ordinary differential equations, has been introduced into continuum mechanics. The components of the internal or hidden state variable vector may be scalars, components of vectors, components of second order tensors, etc. Irradiation, hardening, grain size, chemical reaction, etc., are scalar internal variables. Internal strain, dislocation loop density, etc., are all examples of second order tensors whose components are internal state variables. The internal state variables of this report are irradiation, ϕ , which satisfies the neutron density conservation law, and hardening.

The hardening parameter, χ , that changes with plastic processes and depends upon the whole history of motion, is determined from the postulate rate equation

$$\dot{\chi} = \text{tr}(\mathbf{S}\dot{\mathbf{A}}_p) + \alpha_\chi \text{tr}(\mathbf{S}\dot{\mathbf{A}}_c) + \Gamma_\chi \text{tr}(\mathbf{S}\dot{\mathbf{A}}) + \chi_\phi \dot{\phi} - \gamma_\chi \chi,$$

where \mathbf{S} is the rotated stress tensor, $\dot{\mathbf{A}}$ is the log or true strain rate tensor, $\dot{\mathbf{A}}_c$ is the creep strain rate tensor and $\dot{\mathbf{A}}_p$ is the plastic strain rate tensor. The first term on the right of the above equation is the plastic work function. This is a monotonically increasing function that expresses the irreversibility of the plastic process. In order to have irradiation hardening (softening), the fourth term in the above equation has been added. To allow for annealing, which requires $\dot{\chi}$ to be negative whenever χ is greater than zero, two annealing terms have been added. The classical annealing term, $\gamma_\chi \chi$, is independent of rates, greater than or equal to zero, and normally an increasing function with temperature and stress. The second annealing term accounts for some recovery that takes place during the unloading process. This is accomplished by defining the switch Γ_χ to be positive, whenever $\chi > 0$ and $\text{tr}(\mathbf{S}\dot{\mathbf{A}}) < 0$, otherwise, Γ_χ is zero. The theory has been generalized somewhat by adding a term that depends on creep rate.

This isotropic hardening plasticity theory in conjunction with creep has been incorporated into a computer code called PECTCLAD and has been correlated to annealing tests of 20% cold-worked Type 316 stainless steel and to tensile tests of annealed Type 316 stainless steel cladding. This correlated model is used to accurately predict the tensile, burst, and Fuel-Cladding Transient Testing (FCTT) data for 20% cold-worked Type 316 stainless steel cladding over a wide range of temperatures, temperature rates and strain rate. An excellent comparison is obtained between the predicted and measured data.

1. Introduction

The Hanford Engineering Development Laboratory (HEDL) has been developing a set of analytic methods to reliably predict the performance of Liquid Metal Fast Breeder Reactor (LMFBR) fuel pins during normal, design basis, and hypothetical transients [1]. These fuel pins are composed of uranium plutonium mixed oxide pellets encased in a stainless steel cladding [2]. The cladding is the primary structural member that provides spatial constraints to the pellets, isolates the sodium coolant from the pellets and provides a containment vessel for the fission product gases that are generated by the fission events. The neutron history and distribution and the coolant pressure and temperature histories define the fuel pin environment. This environment provides the fuel pin external loads and reflects normal reactor operation such as startups, shutdowns, power cycles, steady-state operation and off-normal events such as loss-of-coolant transients and reactivity transients. The fuel pin design, must assure that economic goals, exposure goals and cladding integrity can be obtained for a given environment.

Fuel pin cladding design and failure analyses must consider creep rupture, strain-cycle fatigue, local hot spots, fuel-cladding interaction (both mechanical and chemical), fabrication defects and changes in material properties due to irradiation, temperature, carburization, etc. Thus, design criteria which consider these effects must be established. These criteria may be based upon stress (yield or ultimate strength), strain (uniform or total elongation), displacement (to ensure channel clearances), temperature (below coolant boiling), time or a combination of the above, i.e., life fraction, strain fraction, elastic potential energy, etc. To assure that the environment, design, material properties, and criteria are compatible with each other analyses encompassing reactor physics, thermodynamics, metallurgy, and mechanics are necessary.

Fuel cladding behavior modeling is not a simple extension of the experimental and interpretive methods used in classical mechanics. New concepts are needed to describe materials in a reactor environment where temperature, temperature gradient, strain and strain rate are changing rapidly with time. Some aspects of continuum physics used to develop constitutive equations for fuel cladding behavior are presented in this study.

Before we proceed a word about the notation is in order. The direct tensor notation of Truesdell and Noll [3] is used throughout this paper. In particular, scalars are indicated by the Greek alphabet. Points and vectors are denoted by Latin minuscules: x, q, \dots . Second order tensors are indicated by Latin majuscules: T, F, \dots . Second order tensors and linear transformations are regarded as the same. If A is a linear transformation, A^T denotes its transpose, A^{-1} its inverse, $\text{tr}A$ its trace, $\det A$ its determinant and $\hat{A} = A - \frac{1}{3}(\text{tr}A)I$ its deviatoric tensor, where I is the unit tensor. Fourth order tensors are denoted by italic Latin majuscules, \mathcal{K}, \dots .

There are exceptions to the notation rules, e.g., motion. The motion χ relates the spatial coordinates x to the material coordinates X and time t , i.e., $x = \chi(X, t)$. The material gradient of the motion $\nabla_X \chi$ is called the deformation gradient F , i.e., $F(X, t) = \nabla_X \chi(X, t)$. The tensor F has a unique representation as a product of an orthogonal tensor R , i.e., $R^{-1} = R^T$, called the rotation tensor, and a positive definite symmetric tensor U , i.e., $U = U^T$, called the right stretch tensor. We have $F = RU$. The velocity vector \dot{x} is determined by taking the material derivative of the motion, i.e., by taking the derivative of

χ with respect to t , keeping χ fixed. The spatial gradient, denoted by grad , of the velocity vector, $\text{grad } \dot{\chi}$, is related to \underline{F}_κ by $\text{grad } \dot{\chi} = \dot{\underline{F}}_\kappa^{-1} = \dot{\underline{R}}\underline{R}^T + \underline{R}\underline{U}\underline{U}^{-1}\underline{R}^T$.

Coleman and Gurtin [4] have introduced into continuum mechanics the theory of internal state variables. They have studied the thermodynamics of nonlinear materials with internal state variables and characterized the material by five response functions of the type $\psi = \psi(\theta, \underline{F}_\kappa, \underline{g}, \underline{q})$, where θ is the absolute temperature, $\underline{g} = \text{grad } \theta$ is the spatial gradient of temperature, and \underline{q} is the internal state variable vector, whose evolution is governed by ordinary differential equations. The v -components (q_1, q_2, \dots, q_v) of the internal or hidden state variable vector \underline{q} may be scalars, components of vectors, components of second order tensors, etc. Irradiation, hardening, grain size, chemical reaction, etc., are scalar internal variables. Irradiation strain, creep strain, internal strain, dislocation loop density, etc., are all examples of second order tensors whose components are internal state variables.

Bowen [5] is concerned with the formulation of a thermomechanical theory of a mixture of chemically reacting materials without diffusion. He includes the time rate of change of the deformation gradient $\dot{\underline{F}}_\kappa$ as an independent variable in his postulated response functions, i.e., $\psi = \bar{\psi}(\theta, \underline{F}_\kappa, \dot{\underline{F}}_\kappa, \underline{g}, \underline{q})$. He showed that the specific free energy function ψ and hence, the specific entropy η , since $\eta = -\partial_\theta \psi$, are independent of the temperature gradient \underline{g} and $\dot{\underline{F}}_\kappa$ by the general restrictions which the Clausius-Duhem inequality places on response functions. He also showed that $\psi = \bar{\bar{\psi}}(\theta, \underline{\zeta}, \underline{q})$, where $\underline{\zeta} = \underline{F}_\kappa^T \underline{F}_\kappa$ satisfies the axiom of material frame indifference, and is called the right Cauchy-Green tensor.

Recently, Lubliner [6] showed that the non-equilibrium thermodynamics based on internal variables may be used as a foundation on which constitutive laws for solids (viscoelastic, viscoplastic and plastic) may be established. He included the temperature rate $\dot{\theta}$ in his list of argument for the response functions. Kelly and Gillis [7] identified certain internal variables with dislocation densities and the mobile fractions of each kind of dislocation. They used the Gibbs' thermodynamic potential as one of the response functions. Bienick [8] has developed a model of elasto-plastic solids in which the effect of strain hardening is undergoing relaxation or annealing. He included the plastic strain \underline{A}_p in his list of arguments for the response functions. Hahn [9] studied a particular type of internal state variables theory where the differential equations are linear in temperature and deformation rates. He used the multiplicative decomposition of the deformation gradients, i.e., $\underline{F}_\kappa = \underline{F}_{\kappa E} \underline{F}_{\kappa P}$, where $\underline{F}_{\kappa E}$ and $\underline{F}_{\kappa P}$ are the local elastic and plastic deformation tensors, respectively.

This multiplicative decomposition was first studied by Lee and Liu [10]. Recently, Clifton [11] has shown that the two decompositions $\underline{F}_{\kappa E} \underline{F}_{\kappa P}$ and $\underline{F}_{\kappa P} \underline{F}_{\kappa E}$ are equivalent. If we define elastic and plastic strain tensors by $\underline{E}_\kappa = (\underline{F}_{\kappa E}^T \underline{F}_{\kappa E} - \underline{I})/2$, and $\underline{P}_\kappa = (\underline{F}_{\kappa P}^T \underline{F}_{\kappa P} - \underline{I})/2$, then the total engineering strain is the sum of two parts $\underline{E}_\kappa = (\underline{\zeta} - \underline{I})/2 = (\underline{F}_{\kappa E}^T \underline{F}_{\kappa E} - \underline{I})/2$ or $\underline{E}_\kappa = \underline{E}_\kappa + \underline{F}_{\kappa P}^T \underline{E}_\kappa \underline{F}_{\kappa P}$.

Green and Naghdi [12] have introduced similar definitions of strains in their elastic-plastic theory. They define the elastic strain \underline{E}_κ as the differences between the total and plastic parts, i.e., $\underline{E}_\kappa = \underline{E}_\kappa - \underline{P}_\kappa$, hence, by the above definition $\underline{E}_\kappa = \underline{F}_{\kappa P}^T \underline{E}_\kappa \underline{F}_{\kappa P}$, which shows a coupling effect between finite elastic and plastic deformations. The situation becomes more complex when we introduce creep deformation $\underline{F}_{\kappa C}$ to the above decomposition, i.e., $\underline{F}_\kappa = \underline{F}_{\kappa E} \underline{F}_{\kappa P} \underline{F}_{\kappa C}$.

The engineering strain rate tensor $\dot{\epsilon}$ is related to the stretching tensor \dot{D} by

$$\dot{\epsilon} = \dot{F}^T \dot{D} \dot{F} = \dot{U} \dot{U} = (\dot{F}_E^T \dot{\epsilon} + \dot{\epsilon}^T \dot{F}_E) / 2 = \dot{\epsilon}_P + \dot{\epsilon}_{EP}^T \dot{F}_E \dot{F}_E + \dot{F}_E^T \dot{\epsilon}_{EP} \dot{F}_E + \dot{\epsilon}_{EP}^T \dot{F}_E \dot{F}_E + \dot{F}_E^T \dot{\epsilon}_{EP} \dot{F}_E,$$

where the log strain rate tensor \dot{A} is defined by

$$\dot{A} = \dot{R}^T \dot{D} \dot{R} = (\dot{U} \dot{U}^{-1} + \dot{U}^{-1} \dot{U}) / 2. \tag{1}$$

Reiner [12] has stated that when the displacement gradient is finite, the deformations must be measured in terms of the logarithmic measure of strain. When the motion is such that $\dot{U} \dot{U}^{-1} = \dot{U}^{-1} \dot{U}$ (one-dimensional motions in particular) eq. (1) can be integrated to obtain $\dot{A} = \ln \dot{U}$; this shows why \dot{A} is called the log strain tensor. The tensor \dot{A} has been called the co-rotational stretching tensor and is objective to rigid body rotations.

Ting [14] has defined the elastic and plastic strain rates differently from Green and Naghdi; $\dot{\epsilon}_{E*} = \dot{F}_{EP}^T \dot{\epsilon}_{EP} \dot{F}_{EP}$, and $\dot{\epsilon}_{P*} = \dot{\epsilon}_P + \dot{\epsilon}_{EP}^T \dot{F}_{EP} \dot{F}_{EP} + \dot{F}_{EP}^T \dot{\epsilon}_{EP} \dot{F}_{EP}$, so that his elastic stretching tensor $\dot{R}_{E*} = (\dot{F}_E^T)^{-1} \dot{\epsilon}_{E*} \dot{F}_E^{-1} = (\dot{F}_E^T)^{-1} \dot{\epsilon}_{EP} \dot{F}_E^{-1}$, depends only on elastic deformations. It is not clear at this time which strain rate tensor should be used in the formulation of nonelastic deformations. Henceforth, the independent variables of this paper are temperature θ , irradiation ϕ , [15], hardening χ , the co-rotational stretching tensor \dot{A} its inelastic part \dot{A}_I (expansion, swelling, irradiation creep) and its nonelastic part $\dot{A}_N = \dot{A}_C + \dot{A}_P$, where \dot{A}_C and \dot{A}_P are the creep and plastic log strain tensors, respectively. The elastic strain tensor \dot{A}_E , defined by $\dot{A}_E = \dot{A} - \dot{A}_I - \dot{A}_N$, is used whenever possible to reduce equation length. In the special case of one-dimensional motions we can show that the above partitioning of \dot{A} is equivalent to the multiplicative decomposition of $\dot{U} = \dot{U}_{EI} \dot{U}_N = \dot{U}_{EI} \dot{U}_C \dot{U}_P$.

In Section 2, the entropy inequality is used to derive a set of necessary and sufficient conditions that the response function must obey. In the next five sections we show how the theory of internal state variables can be used to introduce irradiation, porosity, inelastic strains, nonelastic strains and hardening into the thermomechanical behavior of materials. A summary of results is presented in Section 8.

2. Thermodynamics and Theory

In the present theory the material at point χ is characterized by eight response functions for the specific free energy ψ , specific entropy η , stress tensor \dot{T} , heating-flux vector \dot{h} , irradiation rate $\dot{\phi}$, hardening rate $\dot{\chi}$, inelastic strain rate \dot{A}_I and nonelastic strain rate \dot{A}_N of the form $\psi = \psi(\theta, \phi, \chi, \dot{A}, \dot{A}_I, \dot{A}_N, \nabla\theta, \dot{\theta}, \dot{\phi}, \dot{\chi}, \dot{A}, \dot{A}_I, \dot{A}_N)$, where $\nabla\theta = \dot{F}_E^T \text{grad}\theta$ is the material gradient of temperature. In this form the differential equations for the internal state variables are implicit, which can be put into standard form by successive substitution, i.e.,

$$\begin{aligned} \dot{\phi} &= \hat{\phi} + \dot{\theta} \partial_{\theta} \phi + \text{tr}(\dot{A} \partial_{\dot{A}} \phi), \\ \dot{\chi} &= \hat{\chi} + \dot{\theta} \partial_{\theta} \chi + \text{tr}(\dot{A} \partial_{\dot{A}} \chi), \\ \dot{A}_I &= \hat{A}_I + \dot{\theta} \partial_{\theta} \dot{A}_I + (\partial_{\dot{A}} \dot{A}_I) \dot{A}, \\ \dot{A}_N &= \hat{A}_N + \dot{\theta} \partial_{\theta} \dot{A}_N + (\partial_{\dot{A}} \dot{A}_N) \dot{A}, \end{aligned} \tag{2}$$

where the coefficients $\hat{\phi}, \partial_{\theta} \phi, \dots, \partial_{\dot{A}} \dot{A}_N$ are independent of rates, i.e., $\hat{\phi} = \hat{\phi}(\theta, \phi, \chi, \dot{A}, \dot{A}_I, \dot{A}_N, \nabla\theta)$, etc. The other four response functions can now be written as

$$\begin{aligned} \psi &= \hat{\psi}(\theta, \phi, \chi, \mathbb{A}, \mathbb{A}_I, \mathbb{A}_N, \nabla\theta, \dot{\theta}, \dot{\mathbb{A}}), \\ \eta &= \hat{\eta}(\theta, \phi, \chi, \mathbb{A}, \mathbb{A}_I, \mathbb{A}_N, \nabla\theta, \dot{\theta}, \dot{\mathbb{A}}), \\ \mathbb{T} &= \hat{\mathbb{T}}(\theta, \phi, \chi, \mathbb{A}, \mathbb{A}_I, \mathbb{A}_N, \nabla\theta, \dot{\theta}, \dot{\mathbb{A}})R^T, \\ \mathbb{h} &= \hat{\mathbb{h}}(\theta, \phi, \chi, \mathbb{A}, \mathbb{A}_I, \mathbb{A}_N, \nabla\theta, \dot{\theta}, \dot{\mathbb{A}}). \end{aligned}$$

These eight equations satisfy the principle of material frame indifference, which state that the response functions are unchanged after a change of frame, and the principle of equipresence [3], which forbids the engineer from choosing independent variables impulsively.

The balance of specific internal energy ϵ is postulated to be

$$\rho \dot{\epsilon} = \text{tr}(\mathbb{T}\mathbb{D}) + \text{div}\mathbb{h} + \rho\gamma, \tag{3}$$

where $\dot{\epsilon}$ is the material derivative of the specific internal energy, $\text{div}\mathbb{h}$ is the spatial divergence of the heating-flux vector \mathbb{h} , γ is called the specific heat absorption and is the heating-supply density, \mathbb{D} is again the stretching tensor and \mathbb{T} is the stress tensor, which measures the contact force per unit area in the deformed configuration.

The mass density ρ is determined from the postulated equation of continuity;

$$\dot{\rho} + \rho \text{div}\dot{\chi} = 0, \tag{4}$$

where $\dot{\rho}$ is the material derivative of the mass density and $\text{div}\dot{\chi}$ is the spatial divergence of the velocity vector. This equation can be rewritten in terms of the trace of the log strain rate $\text{tr}\dot{\mathbb{A}}$ [15]; $\text{tr}\dot{\mathbb{A}} = -\dot{\rho}/\rho$, which can be integrated with respect to time to obtain a relation between the log strain tensor \mathbb{A} and the mass density or specific volume v ;

$\text{tr}\mathbb{A} = \ln(\rho_R/\rho) = \ln(v/v_R)$, where ρ_R and v_R are the reference density and specific volume, respectively. The absolute value of the determinant of the deformation gradient $|\det\mathbb{F}|$, denoted by J , is related to $\text{tr}\mathbb{A}$ by

$$J = |\det\mathbb{F}| = v/v_R = \rho_R/\rho = \exp(\text{tr}\mathbb{A}).$$

The second axiom of thermodynamics is an inequality usually called the Clausius-Duhem inequality and is postulated as

$$\rho \dot{\eta} - \text{div}(h/\theta) - \rho\gamma/\theta \geq 0, \tag{5}$$

where $\dot{\eta}$ is the material derivative of the specific entropy or caloric [16] and $\theta > 0$ is the absolute temperature. This inequality equation and the balance of eq. (3) can be rewritten in terms of the specific free energy; $\psi = \epsilon - \eta\theta$, the co-rotational strain rate $\dot{\mathbb{A}}$ and the rotated stress tensor \mathbb{S} [17], which is objective to rigid body rotations and is related to \mathbb{T} by $\mathbb{S} = R^T \mathbb{T} R$. The principal values of \mathbb{S} are the same as those of \mathbb{T} ; the principal directions of \mathbb{T} are rotated from those of \mathbb{S} by the same amount as the principal direction of strain in the deformed body are totated from the principal directions of strain in the reference configuration. The two rewritten equations are

$$\begin{aligned} \rho(\dot{\psi} + \eta\dot{\theta} + \dot{\eta}\theta) &= \text{tr}(\mathbb{S}\dot{\mathbb{A}}) + \text{div}\mathbb{h} + \rho\gamma, \\ -\rho(\dot{\psi} + \eta\dot{\theta}) + \text{tr}(\mathbb{S}\dot{\mathbb{A}}) + \mathbb{h}\cdot\mathbb{g}/\theta &\geq 0, \end{aligned} \tag{6}$$

where $\mathbb{g} = \text{grad}\theta$. We are now in a position to examine whether or not the response functions are compatible with the reduced dissipation inequality (6)₂.

After we take the material derivatives of the free energy and substitute it into (6)₂,

the Coleman method can be applied to show that the free energy function is independent of the material temperature gradient, temperature rate and log strain rate. Therefore, $\psi = \hat{\psi}(\theta, \phi, \chi, \hat{A}, \hat{A}_I, \hat{A}_N)$ and the inequality, after we substitute for the internal state variable rates, becomes

$$\begin{aligned} & \rho \text{tr} \{ A [\hat{S} / \rho - \partial_{\hat{A}} \hat{\psi} - \partial_{\hat{A}} \phi \partial_{\phi} \hat{\psi} - \partial_{\hat{A}} \chi \partial_{\chi} \hat{\psi} - (\partial_{\hat{A}} \hat{A}_I) \partial_{\hat{A}_I} \hat{\psi} - (\partial_{\hat{A}} \hat{A}_N) \partial_{\hat{A}_N} \hat{\psi}] \} \\ & - \rho \hat{\theta} [\dot{\eta} + \partial_{\theta} \hat{\psi} + \partial_{\theta} \phi \partial_{\phi} \hat{\psi} + \partial_{\theta} \chi \partial_{\chi} \hat{\psi} + \text{tr}(\partial_{\theta} \hat{A}_I \partial_{\hat{A}_I} \hat{\psi}) + \text{tr}(\partial_{\theta} \hat{A}_N \partial_{\hat{A}_N} \hat{\psi})] \\ & - \rho \hat{\phi} \partial_{\phi} \hat{\psi} - \rho \hat{\chi} \partial_{\chi} \hat{\psi} - \rho \text{tr}(\hat{A}_I \partial_{\hat{A}_I} \hat{\psi}) - \rho \text{tr}(\hat{A}_N \partial_{\hat{A}_N} \hat{\psi}) + \hat{h} \cdot \hat{g} / \theta \geq 0. \end{aligned} \quad (7)$$

The fourth order tensors $\partial_{\hat{A}} \hat{A}_I$ and $\partial_{\hat{A}} \hat{A}_N$ are assumed to be symmetric in the above equation.

If we had considered the case when the stress and heating-flux response function are independent of temperature rate, then eq. (7) implies that specific entropy is independent of \hat{A} and $\hat{\theta}$, i.e., $\eta = \hat{\eta}(\theta, \phi, \chi, \hat{A}, \hat{A}_I, \hat{A}_N, \nabla \theta)$, and is determined by

$$\eta = - [\partial_{\theta} \hat{\psi} + \partial_{\theta} \phi \partial_{\phi} \hat{\psi} + \partial_{\theta} \chi \partial_{\chi} \hat{\psi} + \text{tr}(\partial_{\theta} \hat{A}_I \partial_{\hat{A}_I} \hat{\psi}) + \text{tr}(\partial_{\theta} \hat{A}_N \partial_{\hat{A}_N} \hat{\psi})].$$

The classical entropy relation, $\eta = -\partial_{\theta} \hat{\psi}$, is now obtained if all of the internal state variable rates are independent of temperature rate. Also, η becomes independent of the temperature gradient with these assumptions.

Likewise, if $\hat{\mathcal{T}}$, \hat{h} and $\hat{\eta}$ are independent of \hat{A} , then the rotated stress tensor is independent of both \hat{A} and $\hat{\theta}$, i.e., $\hat{\mathcal{S}} = \hat{\mathcal{S}}(\theta, \phi, \chi, \hat{A}, \hat{A}_I, \hat{A}_N, \nabla \theta)$, and is derived to be

$$\hat{\mathcal{S}} = \rho [\partial_{\hat{A}} \hat{\psi} + \partial_{\hat{A}} \phi \partial_{\phi} \hat{\psi} + \partial_{\hat{A}} \chi \partial_{\chi} \hat{\psi} + (\partial_{\hat{A}} \hat{A}_I) \partial_{\hat{A}_I} \hat{\psi} + (\partial_{\hat{A}} \hat{A}_N) \partial_{\hat{A}_N} \hat{\psi}].$$

If both of the above conditions are satisfied, then eq. (7) implies that

$$\hat{\phi} \partial_{\phi} \hat{\psi} + \hat{\chi} \partial_{\chi} \hat{\psi} + \text{tr}(\hat{A}_I \partial_{\hat{A}_I} \hat{\psi}) + \text{tr}(\hat{A}_N \partial_{\hat{A}_N} \hat{\psi}) - \hat{h} \cdot \hat{g} / (\rho \theta) \leq 0. \quad (8)$$

In general eq. (8) can not be resolved into an internal dissipation inequality and a heat-conduction inequality, $\hat{h} \cdot \hat{g} \geq 0$. However, if the internal state variable rates are independent of the temperature gradient, then the heat-conduction inequality is satisfied by postulating the heating-flux vector as $\hat{h} = \hat{k} \cdot \nabla \theta$, since

$$\hat{h} \cdot \hat{g} = \hat{h} \cdot \text{grad} \theta = \hat{h} \cdot (\hat{k}^{-1})^T \nabla \theta = \nabla \theta \cdot \hat{k}^{-1} \hat{h} = \nabla \theta \cdot \hat{k} \nabla \theta \geq 0.$$

The second order tensor \hat{k} is called the thermal conductivity tensor and is semi-positive definite.

3. Irradiation Rate

There are several measures of irradiation. The fuel pin cladding material properties have been correlated to the total fluence nvt , fluence with energies greater than 0.1 MeV, and the product $\bar{E} nvt$, where n is the neutron density function, v is the speed of the neutrons and \bar{E} is the mean kinetic energy. The energy \bar{E} is related to the speed by $\bar{E} = mv^2/2$, where m is the mass of the neutron. In all cases irradiation, denoted by the parameter ϕ , is related to the neutron density, which satisfies the conservation law [18], $\dot{n} = \Gamma_N - \Sigma_A vn$, where Γ_N is the external neutron source rate and Σ_A is the macroscopic absorption cross section. These coefficients are temperature and deformation dependent. However, at this point we will simplify the theory by uncoupling the reactor physics from the thermodynamics and mechanical behavior of the materials. Therefore, we assume that

the irradiation rate $\dot{\phi}$ is a known function of position and time and is independent of temperature, strain, etc., i.e., $\dot{\phi} = \hat{\phi}(\chi, t)$. This equation can be integrated with respect to time to obtain the internal state variable called irradiation.

The irradiation rate coefficients $\partial_{\theta}\hat{\phi}$ and $\partial_{\Lambda}\hat{\phi}$ are now zero and the inequality, $\hat{\phi}\partial_{\phi}\hat{\psi} \leq 0$, must be satisfied. Therefore, $\partial_{\phi}\hat{\psi} \leq 0$, since $\hat{\phi} \geq 0$, for all time. The coefficient $\zeta = -\partial_{\phi}\hat{\psi}$, is part of the gamma heating coefficient, which is always positive or equal to zero. If the elastic moduli are irradiation dependent, then the inequality implies that the moduli decrease with increasing irradiation. This result is supported by the work of Straalsund and Day [19] who found significant decreases in the elastic moduli of irradiated type 304 stainless steel. However, the moduli decrease may be explained by an increase in porosity due to irradiation swelling.

This is but one way to include irradiation effects on structural materials. A far better way has been presented by Dunwoody and Müller [20]. Their work is directed toward a theory of mixtures of chemically reacting constituents where each constituent is allowed its own temperature. They point out that their theory may find an application in mixtures of constituents whose molecules differ largely in mass. Clearly, the condition of very different molecular masses is satisfied in reactor physics where the constituents are neutrons and heavy atoms. This theory of mixtures is beyond the scope of this paper and will not be presented.

4. Porosity

The porosity Π is defined by

$$\Pi = 1 - \bar{\rho}/\rho = 1 - \bar{v}/v, \tag{9}$$

and is a function of both the apparent density $\rho = 1/v$, and the theoretical density $\bar{\rho} = 1/\bar{v}$. Eq. (9) can be obtained by considering the following: Define the apparent volume of a macroelement V , as the sum of the solid volume \bar{V} , with density $\bar{\rho}$, and the cavity volume \hat{V} , with density $\hat{\rho}$, i.e., $V = \bar{V} + \hat{V}$. Volume concentration of particles is defined by $\Pi = \hat{V}/V$. Therefore, $\hat{V} = \Pi V$, and $\bar{V} = (1 - \Pi)V$. The total weight W of the macroelement is $W = \rho V = \bar{\rho} \bar{V} + \hat{\rho} \hat{V}$. By substituting for the solid and cavity volumes, we can show that the apparent density is $\rho = \bar{\rho} (1 - \Pi) + \hat{\rho} \Pi$. Eq. (9) is now obtained by assuming $\hat{\rho} = 0$.

The material derivative of the porosity is $\dot{\Pi} = (1 - \Pi)(\dot{\bar{\rho}}/\bar{\rho} - \dot{\rho}/\rho) = (1 - \Pi)(\dot{\bar{\rho}}/\bar{\rho} + \text{tr}\dot{\Lambda}_E^A)$. We will assume that only elastic deformations occur in a theoretical dense material; i.e., $\dot{\bar{\rho}}/\bar{\rho} = -\text{tr}\dot{\Lambda}_E^A$. Therefore, the porosity rate is determined by $\dot{\Pi} = (1 - \Pi)(\text{tr}\dot{\Lambda} - \text{tr}\dot{\Lambda}_E^A)$, or $\dot{\Pi} = (1 - \Pi)(\text{tr}\dot{\Lambda}_I^A + \text{tr}\dot{\Lambda}_N^A)$, and is independent of elastic strain rates. The porosity is determined by integrating with respect to time,

$$\Pi = 1 - (1 - \Pi_R) \exp[-\text{tr}\dot{\Lambda}_I^A + \dot{\Lambda}_N^A],$$

where Π_R is the reference porosity.

Porosity is not an internal state variable since it is completely defined by knowing the traces of the inelastic and nonelastic strain tensors and the reference porosity.

5. Inelastic Strain Rate

The inelastic strain rate $\dot{\Lambda}_I^A$ is related to the expansion function θ , the irradiations swelling function ϕ , and any other inelastic volume change, such as void growth or collapse,

etc. \hat{A}_I is also related to irradiation creep. Let us define the stress tensor \hat{S}_I by $\hat{S}_I = -\rho \partial_{\hat{A}_I} \hat{\psi}$, then a first order theory of irradiation creep is obtained by postulating $\hat{A}_I = \hat{E}_{II} \hat{S}_I$, where \hat{E}_{II} is a semi-positive definite fourth order irradiation creep coefficient tensor and is function of θ, ϕ, χ , the invariant of strain tensors \hat{A}_0, \hat{A}_I and \hat{A}_N , and $\hat{\phi} = \hat{\phi}(\chi, t)$, i.e., $\hat{E}_{II} = \hat{E}_{II}(\theta, \phi, \chi, \hat{A}_0, \hat{A}_I, \hat{A}_N, \chi, t)$. With these conditions, we have satisfied the inequality

$$-\rho \text{tr}(\hat{A}_I \partial_{\hat{A}_I} \hat{\psi}) = \text{tr}(\hat{A}_I \hat{S}_I) = \text{tr}(\hat{S}_I \hat{E}_{II} \hat{S}_I) \geq 0. \tag{10}$$

The expansion function is defined in terms of the mean linear coefficient of thermal expansion α_θ by $\theta = 3 \ln[1 + \alpha_\theta (\theta - \theta_R)]$, where θ_R is the reference temperature. The theory is completed by defining the coefficient $\partial_{\theta} \hat{A}_I$ in eq. (2)₃ as $\text{tr} \partial_{\theta} \hat{A}_I = \partial_{\theta} \theta$, for an isotropic material. If θ depends on the total strain, then $\text{tr}[(\partial_{\hat{A}_I} \hat{A}_I) \hat{A}_I] = (\hat{A}_I \partial_{\hat{A}_I} \theta)$. However, if θ depends on an internal state variable such as irradiation, then the above postulate for \hat{A}_I must be changed and the inequality (10) may not be satisfied.

The irradiation swelling function ϕ has been treated as an additional expansion function in some cases, i.e., $\hat{A}_I = (\phi \partial_{\phi} \phi) \hat{A}_I + \hat{E}_{II} \hat{S}_I$, and $\text{tr}(\hat{A}_I \hat{S}_I) = (\phi \partial_{\phi} \phi) \text{tr} \hat{S}_I + \text{tr}(\hat{S}_I \hat{E}_{II} \hat{S}_I)$, where the terms $(\phi \partial_{\phi} \phi)$ and $\text{tr}(\hat{S}_I \hat{E}_{II} \hat{S}_I)$ are both positive. However, the trace of the inelastic stress tensor can be positive or negative, therefore, the inequality $\text{tr}(\hat{A}_I \hat{S}_I) \geq 0$, may not be satisfied.

One possible correction that could be made to eliminate the unsatisfied inequality is to recorelate the swelling data in terms of stress, then $\hat{A}_I = \hat{E}_{II} \hat{S}_I$, where \hat{E}_{II} is now a function of a new stress dependent swelling function. One other possible method to eliminate the unsatisfied inequality is to use Dunwoody and Müller theory of mixtures of chemically reacting constituents where each constituent is allowed its own temperature.

6. Nonelastic Strain Rate

The nonelastic strain rate \hat{A}_N is equal to the sum of the creep strain rate \hat{A}_C and the plastic strain rate \hat{A}_P . Let us define the stress tensor \hat{S}_N by $\hat{S}_N = -\rho \partial_{\hat{A}_N} \hat{\psi}$, then a first order theory of thermal creep is obtained by postulating $\hat{A}_C = \hat{E}_{CN} \hat{S}_N$, where \hat{E}_{CN} is a semi-positive definite fourth order thermal creep coefficient tensor and is independent of the temperature rate and the strain rate.

The plastic strain rate is postulated to be proportional to the nonelastic stress tensor \hat{S}_N (assuming a first order theory again) in order to satisfy the entropy inequality. To account for orthotropic material behavior we introduce the plastic stress tensor $\hat{S}_P = \hat{E}_{PN} \hat{S}_N$, where \hat{E}_{PN} is a semi-positive definite fourth order tensor and is a function of both temperature and porosity. The tensor \hat{E}_{PN} relates the shear yield stress ω to the principal components of the tensile yield stress. Also, the trace of the plastic stress tensor is proportional to porosity, $\text{tr} \hat{S}_P = 3 \Pi \alpha_P \text{tr} \hat{S}_N$, where α_P is the plastic dilatation coefficient and is temperature dependent only. Therefore, plastic dilatation occurs only when the porosity is greater than zero, otherwise, plastic incompressibility occurs.

We define $\hat{A}_P = \hat{\lambda}_{PP} \hat{S}_P$, where $\hat{\lambda}_{PP}$ is the plastic unknown coefficient. To determine $\hat{\lambda}_{PP}$ it is necessary to introduce an additional response function. The new response function is called the plastic yield function σ and is postulated to be independent of the temperature rate and the strain rate, i.e., $\sigma = \hat{\sigma}(\theta, \phi, \chi, \hat{A}_0, \hat{A}_I, \hat{A}_N)$. We postulate the plastic yield

function to be $\sigma = 0.5 \text{tr}(\underline{S}_P^2) - \omega^2$, where the shear yield stress ω is an arbitrary function of temperature, irradiation and hardening. The material derivatives of the yield function can be written as $\dot{\sigma} = \dot{\sigma}^* + \text{tr}(\dot{\underline{A}}_P \partial_{\underline{A}} \sigma)$, where $\dot{\sigma}^* = \hat{\sigma} + \hat{\sigma} \partial_{\hat{\sigma}} \sigma + \text{tr}(\dot{\underline{A}} \partial_{\underline{A}} \sigma)$.

In order for the material to plastically deform, it is necessary for the yield function to become a surface, i.e., $\sigma = 0$, which implies $\text{tr}(\underline{S}_P^2) = 2\omega^2$. Also, $\dot{\sigma} = 0$, whenever $\sigma = 0$, which implies $\text{tr}(\dot{\underline{A}}_P \partial_{\underline{A}} \sigma) = -\dot{\sigma}^*$. The plastic strain rate is now completely defined by $\dot{\underline{A}}_P = \dot{\sigma}^* \underline{\Gamma}_P$, where $\underline{\Gamma}_P$ is called the plastic switch and is equal to $-1/\text{tr}(\underline{S}_P \partial_{\underline{A}} \sigma)$, whenever, $\sigma = 0$ and $\dot{\sigma}^* > 0$, otherwise, $\underline{\Gamma}_P$ is zero.

The nonelastic strain rate can now be written as

$$\dot{\underline{A}}_N = \dot{\underline{A}}_C + \dot{\underline{A}}_P = \underline{B}_{CN} \underline{S}_N + \dot{\sigma}^* \underline{\Gamma}_P \underline{S}_P = (\underline{B}_{CN} + \dot{\sigma}^* \underline{\Gamma}_P \underline{B}_{PN}) \underline{S}_N,$$

and satisfies the inequality

$$-\rho \text{tr}(\dot{\underline{A}}_N \partial_{\underline{A}_N} \hat{\psi}) = \text{tr}(\dot{\underline{A}}_N \underline{S}_N) = \text{tr}[\underline{S}_N (\underline{B}_{CN} + \dot{\sigma}^* \underline{\Gamma}_P \underline{B}_{PN}) \underline{S}_N] \geq 0.$$

7. Hardening Rate

To the present theory, we include a hardening parameter χ , that changes with plastic processes and depends on the whole history of motion. Hill [21] has defined the plastic work function as $\chi_P = \int_0^t \text{tr}(\underline{\tau} \underline{D}_P) dt$, where $\underline{\tau}$ is the contact stress tensor and \underline{D}_P is the plastic stretching tensor. This is a monotonically increasing function that expresses the irreversibility of the plastic process. Also, this implies that the elastic moduli decreases with increasing hardening (like the irradiation effect). However, annealing requires the hardening rate to be negative, whenever χ is greater than zero and $\dot{\chi} = -\gamma_\chi \chi$, where γ_χ is the classical annealing coefficient, is normally an increasing function with temperature and stress, and is always positive. The inequality $-\hat{\chi} \partial_{\hat{\chi}} \hat{\psi} = \gamma_\chi \chi \partial_{\hat{\chi}} \hat{\psi} \geq 0$, now requires that $\partial_{\hat{\chi}} \hat{\psi} \geq 0$. Hence, we will assume that the free energy is independent of hardening, i.e., $\partial_{\hat{\chi}} \hat{\psi} = 0$, for all time. This assumption is valid for fuel pin cladding, since large amounts of cold work (20% in particular) does not change the elastic moduli of Type 316 stainless steel [22].

The hardening parameter is determined from the postulate rate equation

$$\dot{\chi} = \text{tr}(\underline{S}_N \dot{\underline{A}}_P) + \alpha_\chi \text{tr}(\underline{S}_N \dot{\underline{A}}_C) + \underline{\Gamma}_\chi \text{tr}(\underline{S}_N \dot{\underline{A}}) + \chi_\phi \dot{\phi} - \gamma_\chi \chi. \tag{11}$$

The first term on the right of eq. (11) is equivalent to the plastic work function of Hill's. The second term generalizes the theory somewhat by depending on creep strain rate. The third term accounts for some recovery that takes place during the unloading process. This is accomplished by defining the hardening switch $\underline{\Gamma}_\chi$ to be positive, whenever $\dot{\chi} > 0$ and $\text{tr}(\underline{S}_N \dot{\underline{A}}) < 0$, otherwise, $\underline{\Gamma}_\chi$ is zero. The fourth term is added to explain irradiation hardening or softening. The hardening coefficient χ_ϕ is a function of the irradiation temperature and is positive for hardening and negative for softening. The last term in eq. (11) is the classical annealing term.

The plastic strain rate and the creep strain rate may be substituted into eq. (11) to obtain the hardening rate standard eq. (2)₂.

8. Summary of Results

In summary, the differential equations for the internal state variables are

$$\dot{\phi} = \dot{\phi}(\chi, t),$$

$$\dot{\chi} = \sigma^* \Gamma_P \text{tr}(S_N \beta_{PN} S_N) + \alpha \chi \text{tr}(S_N \beta_{CN} S_N) + \Gamma_\chi \text{tr}(S_N \hat{A}) + \chi_\phi \dot{\phi} - \gamma_\chi \chi,$$

$$\hat{A}_I = \beta_{II} \hat{\chi}_I + \dot{\theta} \partial_\theta \hat{A}_I,$$

$$\hat{A}_N = (\beta_{CN} + \sigma^* \Gamma_P \beta_{PN}) \hat{\chi}_N.$$

The postulated constitutive equations for the yield stress and the heating-flux vector are

$$\sigma = 0.5 \text{tr}(S_P^2) - \omega^2,$$

$$h = \hat{L} \nabla \theta.$$

A particular free energy function for a first order strain theory is postulated as

$$\rho(\psi - \bar{\psi}) = 0.5 \text{tr}(A_E \beta_{EE} A_E + A_N \beta_{NN} A_N),$$

where the strain independent free energy part $\bar{\psi}$ is defined by

$$\bar{\psi} = \psi_R - \eta_R(\theta - \theta_R) - \bar{\zeta}(\phi - \phi_R) + \int_{\theta_R}^\theta c_\pi d\varepsilon - \theta \int_{\theta_R}^\theta c_\pi d\varepsilon/\varepsilon.$$

The reference coefficients ψ_R , ζ_R , θ_R and ϕ_R are constants. The specific heat at constant pressure c_π is an arbitrary function of temperature. The irradiation coefficient ζ and the semi-positive definite fourth order elasticity tensors β_{EE} and β_{NN} are functions of temperature and irradiation.

The irradiation coefficient ζ , the rotated stress tensors $\hat{\chi}_I$, $\hat{\chi}_N$ and $\hat{\chi}$ and the specific entropy η , are derived from the postulated free energy as

$$\zeta = -\partial_\phi \psi = \bar{\zeta} + \partial_\phi \bar{\zeta}(\phi - \phi_R) - 0.5 \text{vtr}(A_E \partial_\phi \beta_{EE} A_E + A_N \partial_\phi \beta_{NN} A_N),$$

$$\hat{\chi}_I = -\rho \partial_{A_I} \psi = \beta_{EE} A_E,$$

$$\hat{\chi}_N = -\rho \partial_{A_N} \psi = \beta_{EE} A_E - \beta_{NN} A_N,$$

$$\hat{\chi} = \rho \partial_A \psi = \rho(\psi - \bar{\psi})_A + \beta_{EE} A_E,$$

$$\eta = -[\partial_\theta \psi + \text{tr}(\partial_\theta A_I \partial_{A_I} \psi)] = \eta_R + \partial_\theta \bar{\zeta}(\phi - \phi_R) + \int_{\theta_R}^\theta c_\pi d\varepsilon/\varepsilon$$

$$- 0.5 \text{vtr}(A_E \partial_\theta \beta_{EE} A_E + A_N \partial_\theta \beta_{NN} A_N) - \text{vtr}(\partial_\theta A_I \beta_{EE} A_E).$$

The reduced entropy inequality is

$$\hat{\phi}(\zeta + \partial_\phi \phi \text{tr} \hat{\chi}_I) + \text{tr}(S_I \beta_{II} S_I) + \text{tr}[S_N (\beta_{CN} + \sigma^* \Gamma_P \beta_{PN}) S_N] + \nabla \theta \cdot \hat{L} \nabla \theta / \theta \geq 0.$$

This particular theory has been programmed into a computer code called PECTCLAD [23] and is used to analyze LMFBR fuel cladding mechanical behavior under transient conditions. A good correlation to tensile, burst and Fuel Cladding Transient Tester (FCTT) data [24] has been obtained for unirradiated Type 316 stainless steel. The next step in this program is to include irradiation effects.

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10. References

- [1] BARD, F. E., FOX, F. L., WASHBURN, D. F., HANSON, J. E., Analytic Models for Fuel Pin Transient Performance, HEDL-SA-865, Hanford Engineering Development Laboratory, Richland, Washington, September 1976. Presented to the International Meeting on Reactor Safety and Related Physics at Chicago, Illinois (October 5-8, 1976).
- [2] COX, C. M., HANSON, J. E., ROAKE, W. E., SLEMBER, R. J., WEBER, C. E., MILLUNZI, A.C., "FFTF Fuel Pin Design Bases and Performance", Europ. Nucl. Conf., Transactions of the Europ. Nucl. Soc., TANSAO 20 1-820, Amer. Nuc. Soc., Inc., Hinsdale, Illinois (1975).
- [3] TRUESDELL, C., NOLL, W., "The Non-Linear Field Theories of Mechanics," in Encyclopedia of Physics, (S. Flügge, ed.), vol III/3, Springer-Verlag, Berlin (1965).
- [4] COLEMAN, B. D., GURTIN, M. E., "Thermodynamics with Internal State Variables," J. Chem. Phys., vol 47, No. 2, pp. 597-613 (July, 1967).
- [5] BOWEN, R. M., "Thermochemistry of Reacting Materials," J. Chem. Phys., vol 49, No. 4, pp. 1625-1637 (August, 1968).
- [6] LUBLINER, J., "On the Thermodynamic Foundations of Non-Linear Solid Mechanics," J. Non-Linear Mechanics, vol 7, pp. 237-254 (1972).
- [7] KELLY, J. M., GILLIS, P. P., "Thermodynamics and Dislocation Mechanics," J. The Franklin Institute, vol 297, No. 1, pp. 59-74 (January, 1974).
- [8] BIENIEK, M. P., "Annealing Model of Elasto-Plastic Solids," Int. J. Engng. Sci., vol 12, pp. 713-720 (1974).
- [9] HAHN, H. T., "A Finite-Deformation Theory of Plasticity," Int. J. Solids Structures, vol 10, pp. 111-121 (1974).
- [10] LEE, E. H., LIU, D. T., "Finite-Strain Elastic-Plastic Theory with Applications to Plane-Wave Analysis," J. Appl. Phys., vol 38, No. 1, pp. 19-27 (January, 1967).
- [11] CLIFTON, R. J., "On the Equivalence of \bar{F}_{EAP} and $\bar{F}_{P\bar{A}E}$," J. Appl. Mech., pp. 287-289 (March, 1972).
- [12] GREEN, A. E., NAGHDI, P. M., "A General Theory of an Elastic-Plastic Continuum," Arch. Ratl. Mech. Anal., vol 18, pp. 251-281 (1964).
- [13] REINER, M., "Rheology", in Encyclopedia of Physics, (S. Flügge, ed.), vol VI, p. 486, Springer-Verlag, Berlin (1958).
- [14] TING, E. C., "A Thermodynamic Theory for Finite Elastic-Plastic Deformations," Z. Ang. Math. Phys., vol 22/4, pp. 702-713 (1971).
- [15] BARD, F. E., Some Aspects of Continuum Physics Used in Fuel Pin Modeling, HEDL-TME 74-30, Hanford Engineering Development Laboratory, Richland, Washington (June, 1975).
- [16] TRUESDELL, C., Rational Thermodynamics, McGraw-Hill Book Co., New York (1969).
- [17] DILL, E. H., "Simple Materials with Fading Memory," in Continuum Physics, (A. Cemal Eringen, ed.), vol II, Academic Press, New York (1975).
- [18] HARMS, A. A., SCHREINER, S. I., "An Expository Derivation of Neutron Transport Equation," Nuclear Tech., vol 11, pp. 144-149 (May, 1971).
- [19] STRAALSUND, J. L., DAY, C. K., "Effect of Neutron Irradiation on the Elastic Constants of Type 304 Stainless Steel," Nuclear Tech. vol 20, pp. 27-34 (October, 1973).
- [20] DUNWOODY, N. T., MÜLLER, I. "A Thermodynamic Theory of Two Chemically Reacting Ideal Gases with Different Temperatures," Arch. Rat. Mech. Anal., vol 29, pp. 344-369 (1968).
- [21] HILL, R., The Mathematical Theory of Plasticity, Oxford University Press, London (1950).

- [22] Nuclear Systems Materials Handbook, vol 1, Design Data, TID 26666, Hanford Engineering Development Laboratory, Richland, Washington.
- [23] BARD, F. E., GNEITING, B. C., CHRISTENSEN, B. Y., PECTCLAD: A Computer Code to Determine Fuel Pin Cladding Deformations and Stress, HEDL-TME 75-46, Hanford Engineering Development Laboratory, Richland, Washington (December, 1976).
- [24] HUNTER, C. W., FISH, R. L., HOLMES, J. J., "Mechanical Properties of Unirradiated Fast Reactor Cladding During Simulated Overpower Transients," Nuclear Technology, vol 27, pp. 376-380 (November, 1975).