Modelling of the fuel mechanical behavior : from creep laws to internal variable models

Leclercq S.
EDF, France

ABSTRACT : Creep laws such as Bohaboy's one are nowadays commonly used to simulate the fuel pellet response to the sollicitations it faces during its life. These laws are sufficient for describing the base operating conditions (where only creep appears), but they have to be improved for power ramp conditions (where hardening and relaxation appear). The aim of the present paper is to develop a framework in which it could be possible to build models more representative of the fuel pellet in pile sollicitations.

1. INTRODUCTION

The integrity of nuclear fuel rods is one of the major concern for companies like EDF and must be demonstrated, essentially for class I (normal) and II (incidental) operating conditions. As a matter of fact, one potential failure mode of the cladding is the so called Pellet/Cladding Mechanical Interaction which is normally induced by a power ramp. Moreover, the will to extend the fuel's life above four years leads to a need for a better understanding and simulation of the irradiation effects on the fuel mechanical behavior. Thus it clearly appears that the mechanical properties evolution of the cladding, as well as those of the fuel, have to be well known in order to cope with the needs in nuclear fuel rod mechanical modelling, in particular to gain some freedom degrees in nuclear power plants operating conditions. The choice of laws describing such properties remains the major problem.

The problem of determining constitutive equations of any material consists in two steps. First, one has to clearly identify the type of tests that are representative of the real mechanical behavior of the structure. Then the constitutive equations must be written and calibrated on the basis of these experimental results.

The constitutive equations at present existing in the different codes are roughly of the same type. They consist in the writing of a strain rate non linearly depending on the applied stress. Equations such as $\dot{\epsilon} = f(\sigma^n)$ are often used. The exponent of the stress depends on the type of physical mechanism which is involved in the system. Those kinds of constitutive equations are commonly called creep laws, and are able to describe the way the strain increases at constant stress. These constitutive equations provide good predictions of the mechanical behavior of the nuclear fuel rod in normal operating conditions, but are not sufficient to describe the complete mechanical behavior, in particular from the viewpoint of hardening (or softening) properties and relaxation tests. Indeed, even if the creep properties are activated during the normal operating conditions of the nuclear power plant, it is clear that one cannot

assume that the stress remains constant, in particular for ramp conditions. In this case, mechanisms as hardening (or softening) and relaxation are also involved. Usually, the strain-hardening assumption is postulated to solve this problem. Nevertheless, the particular effects associated with relaxation may not be correctly simulated by such correction. Thus one needs constitutive equations that are able to predict such phenomena.

On the other hand, several physical mechanisms arise during the fuel pellet life, that are not *strictly* related to the mechanics, but that have a strong influence on the mechanical behavior of the material during its life. For instance, densification and gas swelling play a major role on the local mass density by changing the porosity inside the material, and hence have an influence on the non-linear rate of viscous deformation. In the actual codes simulating the fuel behavior, these mechanisms are taken into account apart from the mechanical behavior. That is, the deformations induced by densification and gas swelling are parts of the total deformation, but are calculated with no account for the interactions between porosity and viscosity. Thus, the viscoplastic deformation does not depend on the evolution of the porosity, which depends highly on the temperature and the stress field.

Hence, in this case, it seems more accurate to use models that are devoted to this particular behavior, in order to improve the simulation capability of the codes.

In the frame of the Thermodynamics of Irreversible Processes, it is now possible (and of classical use in several fields of mechanical research) to build up models based on the existence of internal variables, able to account for different aspects of the materials mechanical behavior [1]. These methods have already been successfully applied by EDF to simulate the fuel rod Zircaloy cladding behavior [2]. It is proposed here to develop a frame in which such models could be used to describe the elastoviscoplastic behavior of the fuel pellet. In a first part of this paper, the general frame of the model is presented. It is shown how it complies with the second law of thermodynamics. The dependance of the non-linear deformation rate on the effects of swelling and densification is explicitly exhibited. In a second part, it is shown how easy it may be to take into account the irradiation (burnup) on the mechanical behavior thanks to such approach. And the last part of this paper highlights how the parameters of such constitutive equation may be identified. Indeed, this requires also the development of new experimental techniques considering that the integrity loss of the pellet as soon as irradiation proceeds, dismissing the use of standard compressive tests. To this aim, a microindentation machine is developed by TUI (Karlsruhe, Germany) and EDF [3].

2. DESCRIPTION OF THE MODEL

The model has been designed to represent the non-linear mechanical behavior of the fuel pellet, influenced by the effects of swelling and densification that arise at different moments during the life of the fuel rod. The effects of irradiation (i.e. burnup effects) may also be introduced into the present model.

One will use the classical framework of the thermodynamics of irreversible processes to derive the equations described below. In particular, the dissipation aspects will be taken into account on the basis of the Clausius-Duhem inequality, with no direct reference to the classical dissipation potential used in the case of standard materials.

2.1 The general layout

Let us consider a representative volume V of the material in question. As the fuel material (a ceramic obtained by sintering of the Uranium oxide powder) is known to be porous, we define the porosity z as the ratio between the volume of pores V_f and the total volume V . It may change during the fuel's life due to densification (irradiation activated resintering) and gas swelling. Thus, $(1-z)$ constitutes the volume fraction of "sound" material.

It is here assumed that the porosity can have any shape and distribution.

As a conclusion of the layout, one considers that the assumption of small strains is valid here, i.e., the total strain is assumed to be of the form

$$\underline{\varepsilon} = \underline{\varepsilon}^e + \underline{\varepsilon}^{vp}, \quad (1)$$

where $\underline{\varepsilon}^{vp}$ is the viscoplastic (non-linear) part of the deformation. Hence, $\underline{\varepsilon}^e$ is the elastic part of this deformation.

2.2 Thermodynamic principles and constitutive equations

2.2.1 Specific free energy of the system

The Helmholtz specific free energy of the system which can be considered as a two-phase one is written as follows :

$$\Phi(\underline{\varepsilon}^e, z, T) = (1-z)\Phi_1(\underline{\varepsilon}^e, \varepsilon^{vp}, T) + z\Phi_2(\underline{\varepsilon}^e) + \Phi_{it}(z, T), \quad (2)$$

where Φ_1 is the specific free energy of the homogeneous material :

$$\Phi_1 = \frac{1}{2} \underline{\varepsilon}^e \underline{L} \underline{\varepsilon}^e + \Phi^*(\varepsilon^{vp}, T). \quad (3)$$

In this equation, T is the absolute temperature, and Φ^* the free energy correlated with dissipation aspects. $\underline{L}$ is the fourth-order elasticity tensor, while $\underline{\varepsilon}^e$ is the second order elastic deformation tensor.

The term Φ_2 represents the elastic energy stored inside the porosity due to the presence of gas. One assumes for simplicity that it is equal to the elastic energy $(\frac{1}{2} \underline{\varepsilon}^e \underline{L} \underline{\varepsilon}^e)$ stored in Φ_1 .

In equation (2), the term Φ_{it} is called *interaction free energy*. It represents the effects of the porosity upon the total energy of the system. In particular, it takes into account the discontinuities that arise in the strain field, and the relaxations occurring in the stress field when a porosity germinates and grows inside the material.

It is chosen, and this is only an assumption, that Φ_{it} may be of the simple form :

$$\Phi_{it} = z(1-z)F, \quad (6)$$

where F could be a function (experimentally determined) of the burnup BU .

Note that such a non-convex form for the interaction energy has already been used for representing the exotic behavior of shape memory alloys undergoing solid-solid phase transformations [4]. One has to point out that this interaction energy reaches a maximum when $z = 1/2$.

Note also that the free energy of the system is the one of the “sound” material if there is no porosity, and is equal to zero if the volume fraction of porosity is 1 (in this case, there is no more material).

2.2.2 A constitutive assumption

It is now time to give a sense to the viscoplastic strain $\underline{\varepsilon}^{vp}$. In a first approximation, and following the experimental results available in the literature for non-irradiated material [5], the equivalent (in the sense of von Mises) strain rate can be assumed as follows :

$$\dot{\gamma} = \left(\frac{\bar{\sigma}}{K \gamma^{1/M}} \right)^N, \quad (7)$$

where $\dot{\gamma}$ is the rate of equivalent viscoplastic strain, $\bar{\sigma}$ the equivalent stress, and K, M, N are constants. This equation is commonly referred to as the Lemaître law [6].

Thus, the deviatoric part of the viscoplastic strain rate reads :

$$dev(\dot{\underline{\varepsilon}}^{vp}) = \frac{3}{2} \dot{\gamma} \frac{dev \underline{\sigma}}{\bar{\sigma}}. \quad (8)$$

The equation (8) means that the strain rate complies with the normality rule, with possibility of an isotropic hardening, but without any kinematic hardening. This is a restriction which is assumed for this paper, but which can be relaxed in the future.

Let us now deal with the hydrostatic part of the viscoplastic strain. Indeed, in the present case, the volume fraction of porosity may change, but mass conservation is a fundamental principle. Thus, the mass density ρ must change with respect to z . If ρ_0 is the mass density corresponding to z_0 porosity, one can easily derive the following :

$$\frac{1-z}{1-z_0} = \frac{\rho}{\rho_0}. \quad (9)$$

Moreover, neglecting the elastic part of the hydrostatic deformation rate, the equation of mass conservation reads : $\dot{\rho} + 3\rho \dot{\varepsilon}_m^{vp} = 0$, where $\varepsilon_m^{vp} = \frac{1}{3} tr(\underline{\varepsilon}^{vp})$.

This allows one to derive the constitutive equation for the viscoplastic strain rate :

$$\dot{\underline{\varepsilon}}^{vp} = \frac{3}{2} \dot{\gamma} \frac{dev \underline{\sigma}}{\bar{\sigma}} - \frac{1}{3} \frac{\dot{\rho}}{\rho} \underline{I}, \quad (10)$$

where $\underline{I}$ is the second order identity tensor.

2.3 The second principle of thermodynamics

It can be reduced to the well-known Clausius-Duhem inequality, which can be split into the terms of intrinsic dissipation (the mechanical one) and the thermal dissipation. Assuming the classical hypotheses which comply with the thermal inequality, one can write the intrinsic term as follows [6] :

$$\underline{\sigma} \dot{\varepsilon}^{vp} - \rho \frac{\partial \Phi}{\partial z} \dot{z} \geq 0. \quad (11)$$

Dividing (11) by ρ , and using eq. (2) and (10), one obtains :

$$\frac{1}{\rho} \left[\frac{1}{K(\gamma)^{1/M}} \right]^N \bar{\sigma}^{N+1} + \pi^f \dot{z} \geq 0, \quad (12)$$

where $\pi^f = -\Phi^*(\varepsilon^{vp}, T) + F(1-2z) + \frac{1}{\rho_0} \frac{1-z_0}{(1-z)^2} \sigma_m$, and $\sigma_m = \frac{1}{3} \text{tr}(\underline{\sigma})$.

2.4 System evolution and kinetics

One must now write a kinetic equation for z . This equation has to be consistent with the physics of the phenomena, and must also be consistent with equation (12).

From the definition of z , one can easily derive that $\frac{\dot{V}_f}{V_f} = \frac{\dot{z}}{z(1-z)}$, where $\dot{V}_f$ is the volume variation corresponding to the variation of z .

Hence, let us assume

- that there exist a threshold value (zero) for the thermodynamic force π^f that allows the densification and swelling mechanisms (hence the porosity evolution),

- that $\frac{\dot{V}_f}{V_f} = G(\underline{\sigma}, T, BU)$ can be experimentally defined.

One can thus write

$$\dot{z} = H(\Omega)[z(1-z)G], \quad (13)$$

where $\Omega = G\pi^f$. $H(x)$ is the Heavyside function.

This means that gas swelling ($\dot{z} < 0$) cannot proceed while $\pi^f \leq 0$, and densification ($\dot{z} > 0$) occurs only when $\pi^f \geq 0$. The existence of a threshold value for densification and swelling is confirmed by some in pile experimental results.

Note also that the model complies automatically with the second law of thermodynamics thanks to the way the constitutive equations have been written.

3. SIMULATIONS AND DISCUSSIONS

3.1 The functions F and G

The functions F and G play an important role on the global behavior of the material. Indeed, F is responsible for the threshold value for the evolution of the porosity. It is difficult, at present time, to determine experimentally its value. As an example, let us consider the case where $\sigma=0$ at the beginning of the fuel's life. Thus π^f depends only on F (z is equal to z_0). Thus the

evolution of F induces the one of the thermodynamic force associated to z . While π^f is not equal to zero, $\dot{z} = 0$. But as soon as π^f reaches the zero value, the function Ω reaches also zero and $\dot{z}$ can exist, depending on the value of G . That is, F is governing the model in terms of threshold value, while the densification and gas swelling case are governed by G .

Moreover, the function G acts directly on the viscoplastic strain rate. It can be determined experimentally by measurements of the volume variation. Thus, one can obtain the dependence of G on the stress field, the temperature and the burnup. This determination is at present time performed by the CEA (Grenoble, France) in the frame of a cooperation between french companies involved in the nuclear industry.

As an example, one can see on figures 1-2 the effect of this function G on creep simulation tests performed on the material, in the case where gas swelling prevails on densification. Figure 1 shows the influence of the function G ($G = ct/\sigma$) on the creep tests (strain vs time, at $\sigma = 240$ Mpa). Several values of ct are chosen. The corresponding evolution of z is given figure 2. The influence of the porosity growth on the fuel non-linear mechanical properties is obvious, in particular for porosity above 10%, which is commonly reached at the pellet periphery zone.

3.2 The effect of irradiation

3.2.1 From the theoretical viewpoint

Irradiation can be taken into account through different ways. The most natural one is to let F and G depend on the burnup. It is physically what happens because densification and gas swelling depend directly on it. The determination of G by the CEA shall be consistent with this statement.

On the other hand, the irradiation affects not only the evolution of the porosity distribution inside the material, but also the "structural" viscoplastic behavior of the material. Indeed, the material mechanisms that are responsible for viscoplasticity, i.e., grain boundary slipping, creation and migration of dislocations, are altered by irradiation (i.e., fission products, restructuring, secondary compounds,...). Hence, it seems logical to let the parameters (K , M , N) of the deviatoric part of the strain rate depend on the burnup.

The introduction of the burnup in such constitutive equations does not present any difficulty, because the presented frame is open enough.

3.2.2 From the experimental viewpoint

All what has been presented above is valid whatever the material is irradiated or not. The identification of the constitutive parameters is quite easy on non-irradiated material, because specimens of good quality are available. The problem is not so simple for irradiated material, because it becomes fractured, and one cannot use any specimen to perform classical macroscopic tests. Thus, it has been decided to develop other experimental techniques to provide tests useful for the parameter identification.

To this aim, EDF and the Institute for Transuranium Elements of Karlsruhe (Germany) develop together a microindentation technique [3]. A device has been designed and built. It will allow to perform mechanical tests on small irradiated specimens. Rational curves σ, ϵ (at

constant strain rate), creep and relaxation tests will thus help one to improve the constitutive equation relative to the deviatoric part of the strain rate. For instance, a kinematic hardening could be added, or one could take into account restauration effects.

Nevertheless, the experimental results available do not fully correspond to those obtained from classical mechanical tests. This is due to the fact that the stress state induced by the microindenter is highly non-homogeneous. Thus, it is necessary to develop an original identification method devoted to this type of tests. An inverse method coupled with a finite element calculation shall provide the solution desired.

Fig.1 : Creep test

Fig.2 : Porosity evolution

4. CONCLUSIONS

The main objective of the present paper was to clearly exhibit the important role of the porosity on the fuel mechanical behavior. It has been shown that viscoplastic properties are activated by the evolution of the porosity. A general framework has been developed, in agreement with the principles of thermodynamics of irreversible processes. The major result of the present model concerns the fact that the viscoplastic strain is non-deviatoric, due to the porosity growth. The purely deviatoric part of the non-linear strain is taken as the Lemaître law, but any other classical equation may be used. As concerns the hydrostatic part, it is derived from simple assumptions. The coupling between the volume fraction of porosity and the mechanical stress field is introduced into the dissipation term.

The problem is now to clearly identify the functions F and G , in order to establish how the porosity evolves with respect to the mechanic and physic conditions. Mechanical tests on irradiated material, in particular in the so-called rim zone, shall provide numerous informations in a near future. Thus, the next step will be to implement such a model into the existing numerical codes devoted to the fuel rod, in order to improve their capabilities as regards the mechanical simulation of the fuel pellet behavior.

REFERENCES

1. Rice, J.R. 1971. Inelastic constitutive relations for solids : an internal-variable theory and its application to metal plasticity, *J. Mech. Phys. Solids*, **19**, 433-455.
2. Delobelle, P., Robinet, P., Geyer, P., Bouffieux, P., Le Pichon, I. 1995. A unified model to describe the anisotropic viscoplastic behavior of zircaloy-4 cladding tubes, *proc. 11th Symposium « zirconium in the nuclear industry »*, ASTM STP, Garmish-Partenkirchen.
3. Baron, D., Leclercq, S., Spino, J., Taheri, S. 1996. Development of a microindentation technique to determining the fuel mechanical behaviour at high burnup, *IAEA Technical Committee Meeting on Advances in Pellet Technology for improved Performance and High Burnup. Tokyo, 26 October-1 November*.
4. Leclercq, S., Lexcellent, C. 1996. A general macroscopic description of the thermomechanical behavior of shape memory alloys, *J. Mech. Phys. Solids*, **44**, 6, 953-980.
5. Olander, D.O. 1976. *Fundamental aspects of nuclear reactor fuel elements*, Technical Information Center, Office of Public Affairs, Energy Research and Development Administration, Berkeley.
6. Lemaître, J., Chaboche, J.L. 1985. *Mécanique des Matériaux Solides*, Dunod, Paris.

ACKNOWLEDGEMENT

The author wish to thank Dr G. Rousselier for the scientific discussions that initiated this work, and D. Baron and J.M. Ruck for their fruitful remarks.