Modeling of the fuel mechanical behavior : Internal variables and physics of solids

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Abstract

In this paper, one first recalls the issues of the Pellet Cladding Mechanical Interaction (PCMI) occurring in a PWR fuel rod under nominal and incidental operating conditions. In order to demonstrate that PCMI does not induce any fuel rod failure, Electricité de France (EDF) develops constitutive equations allowing to simulate the viscoplastic behavior of the Zircaloy cladding and the UO_2 fuel pellet. The latter is being discussed in the present study. After having identified the mechanisms responsible for the mechanical behavior of industrial pellets, one puts the emphasis on a dislocation based model allowing to describe the non monotonous viscoplastic flow (yield compressive point). This approach is afterwards introduced at a local scale in the framework of a polycrystalline approach. The numerical results exhibit a very good agreement with the experimental ones.

1 Introduction

The fuel rods in Pressurized Water Reactor are made of UO_2 pellets inside a Zircaloy cladding tube. During class 2 incidents or power changes due to load follow, Pellet-Cladding Mechanical Interaction may occur, inducing a cladding stress increase. The viscoplastic behavior of the pellet at its center (high temperature zone) tends to reduce the stress level.

The accurate estimate of the stress level in the cladding is a major concern for EDF since it is the first barrier for irradiation wastes. Therefore in order to quantify sharply the stress level in the cladding EDF R&D develops viscoplastic models. Since PCMI is a transient phenomenon, steady state creep laws are useless, thus mechanical constitutive equations are required. A phenomenological approach was first used, it consisted in a Lemaitre's model extended by (Leclercq, 1998) and (Sauter and Letaief, 2000) to take into account the irradiation effects and the variations of the porosity. These models are unfortunately unable to describe the non monotonous behavior of industrial pellets - i.e. a yield point during strain hardening tests (see Fig. 3a, 3c, 3d) or a sigma creep curve - furthermore their predictive abilities are relatively poor.

Consequently, EDF R&D has begun the development of a thermodynamical approach - the Cunat's Distribution of Non Linear Relaxations (D.N.L.R.) model (see Sauter et al. (2000) or Cunat (1991)). More recently the emphasis has been put on a dislocation based model in the framework of Pilvin and Cailletaud's polycrystalline approach. Hereafter we will focus on the latter.

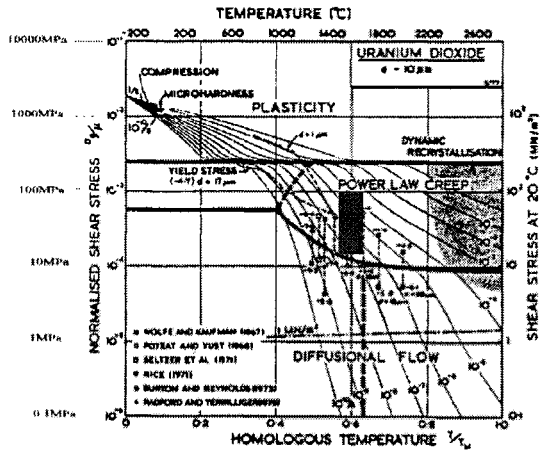
The demonstration requires a recall of the material properties (crystallography and viscoplasticity) and a study of the dominant deformation mechanism (dislocations glide, vacancy diffusion) and damage. Afterwards, the constitutive equations of the model will be described.

2 Main Features of the material

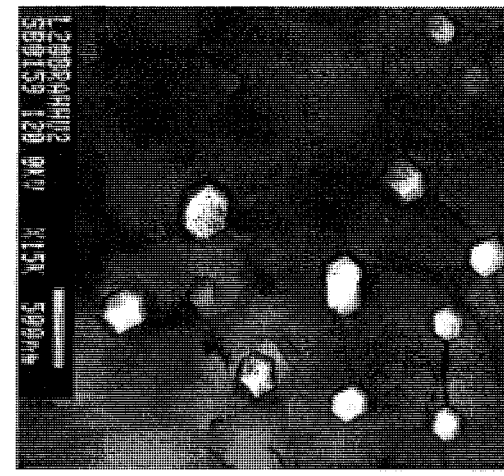
2.1 Crystal properties

UO_2 fuel pellets are partially ionic solids with a fluorite structure. The oxygen ions are arrayed in a simple cubic lattice and the uranium ions form a fcc sublattice. The unoccupied interstitial positions in the body centers of the small cubes do not contain uranium ions (Olander, 1976). At room temperature, they exhibit a very high yield stress and a mainly brittle behavior. This is due to the very high Peierls Nabarro stress $(10^{-2}\mu)$[1] and the very low dislocation density. At high temperatures, viscoplasticity resulting from dislocations motions appears in the range of stress we deal with. The Burgers' vector is $\frac{a_0}{\sqrt{2}}\langle 110\rangle$ where $a_0 = 5.47 * 10^{-10} m$. The easier active slip plane is $\{001\}$ and for higher energy level $\{011\}$

[1] μ is the shear modulus



(a) Deformation-map mechanisms for UO_2 (Ashby and Frost, 1982)



(b) Dislocations pinned on pores

Figure 1: Viscoplastic mechanism and interactions with pores.

and finally $\{111\}$. Note that this is the opposite rule as in crystalline materials. Sometimes, the plane $\{111\}$ interacts with the $\langle 110 \rangle \{001\}$ slip thus an hexagonal network appears (Alamo et al., 1978). As the stacking fault energy is very high, glissile dislocations do not dissociate (see Lefebvre (1976)).

2.2 Viscoplastic properties

Steady state creep of UO_2 has been formerly widely studied. Ashby and Frost (1982) have summarized these results on a deformation-map mechanisms which gives the viscoplastic strain rate versus temperature and the associated deformation mechanism, see Fig.(1a). The colored area stands for the range of stress and temperature of the present study. It brings out that dislocation glide and vacancy diffusion (especially grain boundary diffusion) are likely to occur but actually dislocation glide is the leading viscoplastic deformation mechanism.

On Fig.(1b) one can see dislocations pinned on pores. This mechanism leads the stress necessary to activate dislocation glide to increase. This feature may be responsible for the yield point or the sigma creep curve. These primary stages correspond to a stiff creation of mobile dislocations. Thus, the strain is likely to decrease at the beginning of viscoplastic flow whereas the strain increases. This characteristic has been successfully modeled by Alexander and Haasen (1968), in a framework initially developed for single crystal of silicium which exhibit a high yield point. This will be detailed in the next section.

As UO_2 is a ceramic it is well known that the stoichiometric ratio is likely to evolve if the partial pressure of oxygen is not perfectly controlled. These evolutions have been modeled by Lindemer and Besmann (1985) see Tab.(1) with x the stoichiometric excess. Obviously, the crystallography is closely dependent on the stoichiometry. The atoms of oxygen are in interstitial position so in case of hyperstoichiometry some of these atoms can pin dislocations and trigger a yield point. The literature provides also indications on the influence of stoichiometry on the elastic and viscoplastic properties of UO_2. For a complete review of these dependency (see Sauter (2001)).

2.3 Damage evidences

On Fig.(2), one can hardly see the grain boundary sliding whereas the sample has been deformed to 6% but large cavities have obviously grown in the grain boundary area. This is characteristic of the appearance of damage associated to viscoplasticity. A way to quantitatively estimate the damage is to use a microacoustic method. The evolution of the Young's modulus is estimated by measuring the speed of sound in a thin layer close to the surface of the sample. This evolution is reported in Tab.(2). These values indicate that damage must not be discarded in the modeling of the thermomechanical behavior

Composition	Expression of the oxygen potential $RT \ln p_{O_2}$ (J/mol)
UO_{2-x}	$-1300000 + 225.7T - 3RT \ln \left[\frac{1.5x}{(1+x)^{\frac{2}{3}} \left(1-\frac{x}{2}\right)^{\frac{1}{3}}} \right]$
$UO_{2.00}$ $(T < 3015\,\mathrm{K})$	$-897000 + 224.8T$
UO_{2+x}	
$0 < x < 0.01$	$-360000 + 214T + 4RT \ln \left[\frac{2x(1-2x)}{(1-4x)^2} \right]$
$x > 0.01$	$-312800 + 126T + 2RT \ln \left[\frac{x(1-2x)^2}{(1-3x)^3} \right]$

Table 1: Evolution of the oxygen potential versus x (Lindemer and Besmann, 1985).

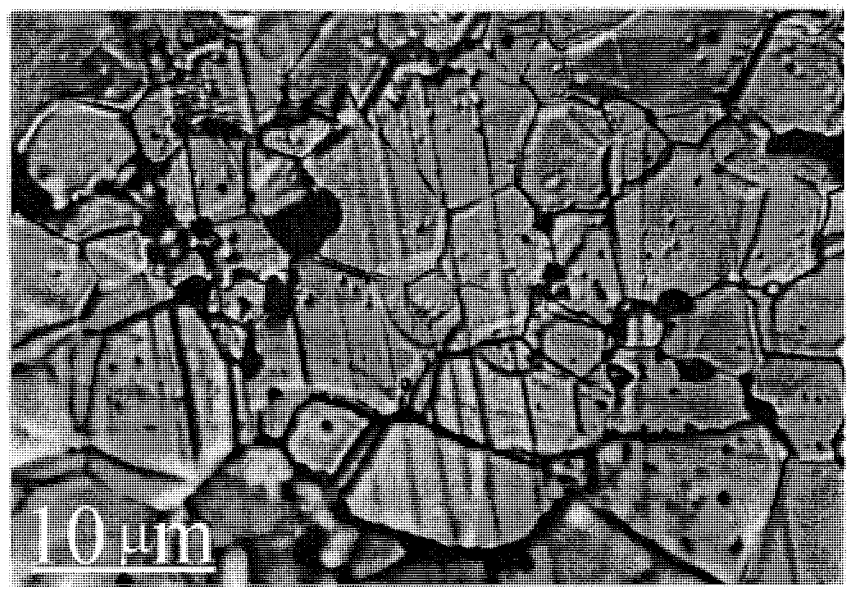


Figure 2: Creation of intergranular cavities in UO_2, $\varepsilon \approx 6\%$ (Vivant-Duguay, 1998)

of UO_2 pellets. Note that damage had never been introduced in the behavior of non irradiated pellets before due to a lack of experimental evidence.

As a first approximation, the Young's modulus will be calculated as decreasing function of the viscoplastic strain ε^{vp}:

$$E^u = E_0^u * (1 - c_d \varepsilon^{vp}) . \tag{1}$$

3 Model description

The Alexander and Haasen's model is well known for its ability to describe the yield point. We first have modified the model according to Kubin and identified its parameters to fit the mechanical behavior of UO_2. Then it has been introduced in the framework of Pilvin and Cailletaud's polycrystalline approach (Cailletaud (1987); Pilvin (1990)) to take into account the crystallographical properties of UO_2 (such as critical resolved shear stress and slip plane orientation) and hence have a better estimation of the internal variables responsible for the viscoplastic behavior of UO_2, and of internal stresses.

Strain (%)	**Young's modulus (MPa)**
0	229000
4.47	163948
6.50	163156
7.02	157663

Table 2: Estimation of the evolution of the Young's modulus due to damage.

This framework is based on a local behavior description (here Alexander and Haasen's approach) at the phase scale (a phase is defined as the continuum media which exhibits one crystalline orientation and a given glide system) and a scale transition which allows to link the local mechanical fields (local stress, local strain) with the macroscopic ones (strain and stress fields at the Representative Elementary Volume scale). Thus a REV is made of several phases (200, with random crystallographic orientation, in order to account for the isotropic behavior of polycrystalline UO_2).

3.1 Local behavior

The Alexander and Haasen's basic model reads:

$$v = v_0 \left(\frac{\tau^*}{\tau_0}\right)^m \exp\left[-\frac{\Delta H_0}{RT}\right], \tag{2}$$

$$\dot{\gamma} = \frac{\dot{\tau}}{M} + \mathsf{b}\rho v, \tag{3}$$

$$\tau^* = \max\left(0; \tau - \tau_i\right), \tag{4}$$

$$\dot{\rho} = K\rho v \tau^* \tag{5}$$

where γ, v, b, ΔH_0, ρ, τ^*, τ_i, M are respectively the shear strain, the speed of dislocations, the magnitude of the Burgers' vector, the activation free enthalpy, the density of dislocations, the effective shear stress, the internal shear stress and the stiffness of the assembly which is equivalent to an apparent shear modulus. Taking into account the stiffness of the experimental device and the range of temperatures M is quite different from the shear modulus μ ($M \simeq \frac{\mu}{10}$ from our identifications which agrees with Rabier and George (1987)'s indications). Furthermore, it is found in the literature $1 \leqslant m \leqslant 2$. Note that the lower value stands for thermodynamical stability.

This model is able to describe the yield point in the case of a strain rate controled test. Indeed, $\tau^* = 0$ until $\tau \leq \tau_i$,hence τ increases and no dislocation is created. As soon as τ yields the value of the internal shear stress (close to the yield point) $\tau^* > 0$ and the multiplication of dislocations occurs (ρ increases). In order to comply with Equation (3), $\dot{\tau}$ must be negative (this transient effect is called the overshoot). Afterwards hardening occurs and the viscoplastic flow becomes monotonous, as an equilibrium arises between the effective shear stress and the dislocation density. According to Alexander and Haasen the evolutions of the yield point τ_{YP} can be estimated by:

$$\tau_{YP} \propto \dot{\gamma}^{\frac{1}{m+2}} \exp\left(\frac{\Delta H_0}{(m+2)\,RT}\right). \tag{6}$$

In order to improve the Alexander and Haasen's basic model, Estrin and Kubin (1986) have proposed kinetics for the dislocation density based on coupled differential equations in which sessile (or forest) dislocations $Y_s = \frac{\rho_f}{\rho_{fo}}$ and glissile (or mobile) dislocations $X_s = \frac{\rho_m}{\rho_{mo}}$ are taken into account.

$$\frac{dX_s}{d\gamma} = q\left(-c_1\sqrt{Y_s} - c_3 X_s + c_4 \frac{Y_s}{X_s}\right) \tag{7}$$

$$\frac{dY_s}{d\gamma} = \left(c_1\sqrt{Y_s} - c_2 Y_s + c_3 X_s\right), \tag{8}$$

.

The Alexander and Haasen's model provides a local behavior in terms of shear strain and stress, for a given glide system. Nevertheless, one has now to evaluate the total viscoplastic strain tensor $\boldsymbol{\varepsilon}^{vp}$. The heterogeneity of the representative element volume (REV) of the material is introduced in the model. Dislocations can not be represented directly in this approach. The first level of heterogeneity is the grain. The morphology of the grains (shape, size) is identical but the grains differ from each other in terms of crystallographic orientations. Thus a phase will be defined with its Euler's angles and its volume fraction f_g which is related to the number of occurrence of an orientation. This set of values defines a FDOC (Function of Distribution of the Crystalline Orientations). From a mechanical point of view, each phase is considered as a continuum media which behavior takes into account the crystallographic structure (fcc) and the glide systems ($\langle 011\rangle(100), \langle 01\bar{1}\rangle 1(01), \langle 01\bar{1}\rangle(111)$). The following variables ($\boldsymbol{\sigma}, \boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}^{vp}$) are average quantities significant of the behavior of any equivalent glide system in the REV. Each system s

is defined by the Schmid's tensor $\boldsymbol{m}_s$ calculated with the direction of glide $\boldsymbol{l}_s$ and the normal vector to the glide plane $\boldsymbol{n}_s$ as follows:

$$\boldsymbol{m}_s = \frac{1}{2}\left(\boldsymbol{l}_s \otimes \boldsymbol{n}_s + \boldsymbol{n}_s \otimes \boldsymbol{l}_s\right), \boldsymbol{l}_s = \frac{\boldsymbol{b}_s}{\mathsf{b}_s}. \tag{9}$$

The contribution of a glide system (s) to the plastic strain $\boldsymbol{\varepsilon}^{vp}$ is given by:

$$\dot{\boldsymbol{\varepsilon}}^{vp} = \sum_{s \in S} \boldsymbol{m}_s \dot{\gamma}_s. \tag{10}$$

Note that we respect the hypothesis of small perturbations hence $\dot{\boldsymbol{m}}_s = \boldsymbol{0}$.
The viscoplastic shear strain rate $\dot{\gamma}_s$ is settled by[2]:

$$\dot{\gamma}_s = \zeta \rho_m \left(\frac{\tau_v}{\tau_i}\right)^m sgn\left(\tau_s\right), \tag{11}$$

$$\tau_v = \max\left(0; |\tau_s| - \tau_i\right), \tag{12}$$

with the following internal stress:

$$\tau_i = \tau_0 + \alpha \mu \mathsf{b} \sqrt{\sum_{r \in S} A_{rs} \rho_r}, \tag{13}$$

where τ_0 is the critical resolved shear stress and ρ_s the total dislocation density ($\rho_s = \rho_f^s + \rho_m^s$). The hardening matrix $\boldsymbol{A}$ can be written $A_{rs} = h_{lat} + (1 - h_{lat})\,\delta_{rs}$ where δ_{rs} is the Kronecker's index so that with $h_{lat} = 1$ one obtains the unity matrix and Taylor's formula can be derived (Franciosi, 1984):

$$\tau_i = \tau_0 + \alpha \mu \mathsf{b} \sqrt{\sum_{r \in S} \rho_r}. \tag{14}$$

With $h_{lat} = 0$, there is no interaction between glide systems. Strain hardening in a given system is controlled by its own density of dislocations. As we do not have any indication on the phenomenon h_{lat} is defined as a variable to be numerically identified. According to Moulin et al. (1999), the value of α is given by:

$$\alpha = \frac{1.48}{2\pi\left(1 - \nu\right)}, \tag{15}$$

$2\pi\left(1 - \nu\right)$ being the signature of edge dislocations.

3.2 Scale transitions

Now that we dispose of the mechanical variables $(\boldsymbol{\sigma}, \boldsymbol{\varepsilon}, \boldsymbol{\varepsilon}^p)$ for each phase, we have to perform a scale transition to obtain the macroscopic data $(\boldsymbol{\Sigma}, \boldsymbol{E}, \boldsymbol{E}^p)$. To the aim, spatial average on the REV are necessary. The REV is defined a set a phases characterized by the FOC:

$$\boldsymbol{\Sigma} = \sum_{g \in G} \left(f_g \boldsymbol{\sigma}\right), \tag{16}$$

$$\boldsymbol{E} = \sum_{g \in G} \left(f_g \boldsymbol{\varepsilon}\right). \tag{17}$$

Hill has written, in case of an elastic-plastic behavior, the scale transition rule as follows:

$$\dot{\boldsymbol{\sigma}} = \dot{\boldsymbol{\Sigma}} + \mathcal{L}^* \left(\dot{\boldsymbol{E}}^p - \dot{\boldsymbol{\varepsilon}}^p\right), \tag{18}$$

where $\mathcal{L}^*$ is a fourth order tensor. This calculus is complex since Eq.(18) is an implicit equation. Solving this equation in case of elastoviscoplasticy does not meet industrial considerations.

[2] ζ, m and ρ_m are material parameters

m	c_1	c_2	c_3	c_4	$\rho_{f_0}\ (m^{-2})$
10.24	9.157	$3.000E3$	$3.144E-7$	$2.987E3$	$2.27*10^6$
c_d	$\zeta\ (s^{-1})$	h_{lat}	Δ	δ	$E_0^u\ (MPa)$
$8.507E-3$	$2.707E-24$	$6.874E-1$	$1.019E4$	$7.941E-3$	$2.023E5$

Table 3: Parameters of the polycrystalline model.

Here the scale transition rule is set *a priori* –explicitly– taking into account the morphologic similarity of grains considered identical with an ellipsoidal shape. This is Pilvin and Cailletaud's method who introduced a set of variables $\boldsymbol{\beta}_g$ taking into account intergranular hardening. the kinetic of which is given phenomenologically Eq.(20):

$$\boldsymbol{\sigma} = \boldsymbol{\Sigma} + \mathcal{C}\left(\boldsymbol{I} - \mathcal{S}\right)\left(\boldsymbol{B} - \boldsymbol{\beta}_g\right), \tag{19}$$

$$\dot{\boldsymbol{\beta}}_g = \dot{\boldsymbol{\varepsilon}}^p - \Delta\left(\boldsymbol{\beta}_g - \delta\boldsymbol{\varepsilon}^p\right)\|\dot{\boldsymbol{\varepsilon}}^p\| \tag{20}$$

where $\boldsymbol{B} = \sum_{g\in G}\left(f_g\boldsymbol{\beta}_g\right)$, $\mathcal{C}$: tensor of elastic moduli (isotropic and homogeneous), $\mathcal{S}$: the Eshelby's tensor for an ellipsoidal shape inclusion. Thus there are new set of parameters to identify (Δ, δ).

According to Pilvin and Geyer (1997), the validity of the scale transition rule, i.e. the values of (Δ, δ), can be checked comparing the stress tensors of particular grains to the ones calculated either with the so-called Berveiller and Zaoui (1979)'s method or using Finite Elements Method. This work is currently in progress.

3.3 Results and discussion

The table 3 provides the set of parameters of the polycristalline model identified on three hardening-relaxation tests presented on Fig. 3. Note first that h_{lat} is between 0 and 1 which means there are some interaction between the slip systems and second Δ and δ are such that the contribution of intergranular phenomena to the strain hardening is quite small. One can see the very good agreement between numerical and experimental tests.

Note that the temperature range of the present study is $[950°C, 1150°C]$, so that the damage due to grain boundary sliding evidenced at temperatures above $400°C$ can be neglected.

4 Conclusion

The aim of the present work was to propose an original approach to simulate the non monotonous viscoplastic behavior of nuclear fuel pellets under nominal and incidental operating conditions.

The leading viscoplastic mechanism of UO_2 pellets has been identified, and a dislocation based approach has been used to represent it at a local scale. The Alexander and Haasen's model was chosen for its ability to reproduce the yield point evidenced on strain rate controlled compressive tests. The use of a polycristalline framework allows one to take into account the crystallographic structure and the grain heterogeneity of UO_2. One can therefore expect good predictive abilities of the present model. The grain boundary sliding which has been evidenced for tests performed above $1400°C$ has been neglected here, as the database available for the identification procedure belongs to $[950°C, 1150°C]$ temperature range. The effect of damage on the elastic properties is taken into account. Nevertheless, future works will manage an extension of the model validity up to $1400°C$, and a damage variable will be introduced in the modeling.

Acknowledgement 1 *The authors would like to thank Dr. A. Mocellin, Dr. F. Dherbey and Mr. M. Sladkoff (CEA) for providing the experimental data. The help of Pr P. Pilvin (Ecole Centrale de Paris) is also gratefully acknowledged.*

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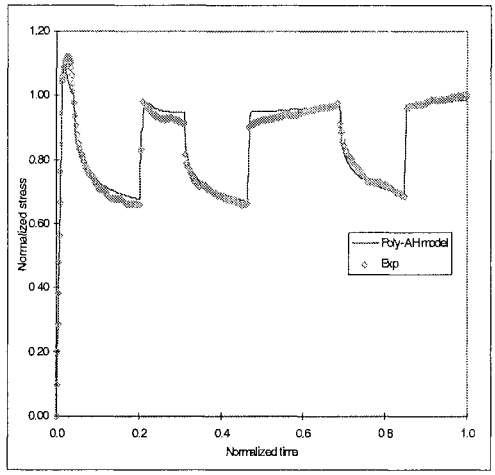
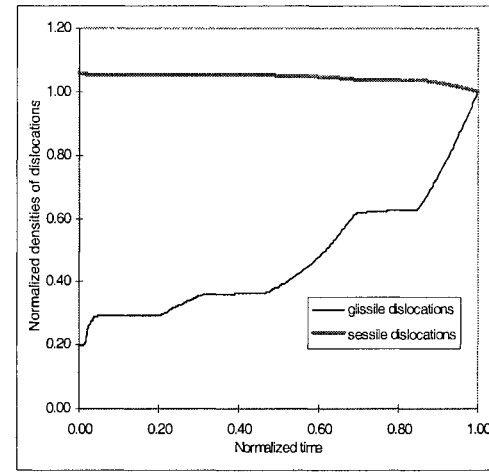
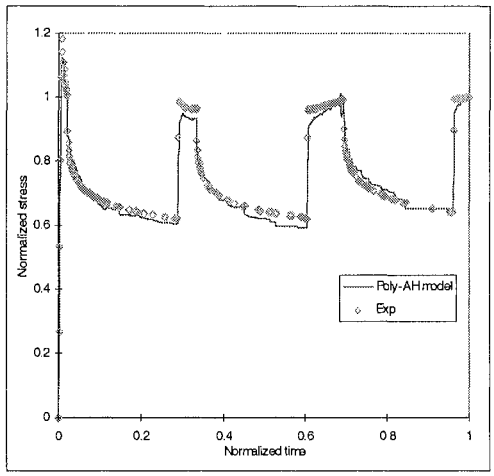


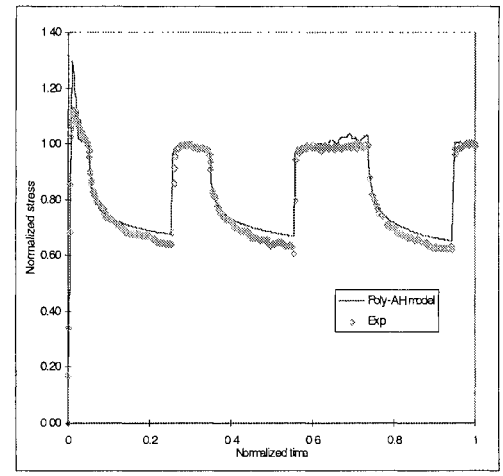
(a) v282: $T = 950C$, $\dot{\varepsilon} = 1.45E-5\,\mathrm{s}^{-1}$



(b) v282: Evolutions of the densities of dislocations



(c) v283: $T = 950C$, $\dot{\varepsilon} = 6.24E-5\,\mathrm{s}^{-1}$



(d) v285: $T = 1150C$, $\dot{\varepsilon} = 2.36E-5\,\mathrm{s}^{-1}$

Figure 3: Identification of the modified Alexander and Haasen's model with two densities of dislocations in the polycrystalline framework, $\varepsilon_M \approx 6.5\%$.

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