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Thermal Creep of MOX Fuel: a Review of Correlations

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ABSTRACT

Demonstrators and prototypes of most promising fast reactors in Europe will be fuelled with MOX at least during their early phase of deployment. The use of MOX fuel makes licensing procedures of innovative systems less complex thanks to a sound knowledge gained on in-pile MOX performance. In this view, operating experience of Phénix and Superphénix reactors is of upmost relevance. Fuel creep strain rate is a relevant quantity for fuel rod performance under Pellet-Cladding Mechanical Interaction conditions (PCMI). Experimental findings have confirmed that, beside temperature, stress, and grain size, other quantities such as porosity, plutonium content, and stoichiometry affect fuel creep. This paper presents a review of experimental findings and correlations published in the open literature. The discussion moves from values of creep rate predicted under reference conditions. Then, each input variable has been varied while maintaining the other ones at their initial value, the creep rate has been re-calculated after that. This approach has allowed to give useful indications concerning relevance and modelling of each input parameter. Results of our analysis showed that porosity and deviation from stoichiometry are sources of discrepancies between models' predictions.

1 INTRODUCTION

Prototypes and demonstrators of most promising innovative fast reactors in Europe will be loaded with MOX fuel having concentrations of plutonium dioxide up to 30 mol% [1]. MOX fuel undergoes a significant restructuring under foreseen irradiation conditions [2]. Recent studies carried out on low-content plutonium MOX for LWR showed, relying on a physics-based approach, that restructuring affects fuel creep behaviour [3,4]. Benchmark and comparisons between fuel performance codes on FBR experimental tests have shown that modelling of fuel creep may partly explain the deviations noted in calculations [5,6]. Models used in fuel performance codes and published in the open literature are mostly based on empirical correlations pointing out that advancements in physics-based approaches could represent an interesting opportunity for future developments. In this paper empirical creep correlations are compared within domains of input variables complying with FBR conditions. Our discussion is focused on the secondary thermal creep without considering the contribution of fission-induced creep.

2 DOMAINS OF INPUT VARIABLES

Correlations of thermal creep rate for MOX fuel depend on: temperature, stress, grain size, porosity, plutonium concentration, O/M ratio. Effect of burn-up should be taken into

account as done for fuel thermal conductivity. Burn-up does not appear among the input variables of correlations discussed here. However, part of the phenomena occurring under irradiation is accounted for through porosity, O/M ratio, plutonium concentration. As mentioned above, similar comments may hold for the heterogeneity of fuel. Domains considered for input variables are resumed in Tab. 1. These indications have been judged to be well representative of conditions occurring under a typical FBR irradiation. The dependence of creep rate on stress is expressed through a polynomial composed of a linear term with stress exponent equal to 1 (diffusion) and a term where the stress is raised to the power either 4.4 or 4.5 (dislocation). Temperature affects thermal creep according to Arrhenius-type functions where values of activation energy depend on corresponding driving process (diffusion, dislocation).

Table 1: Domains of creep input variables encountered in FBR

Input variable	**Domain**
Stress (MPa)	1-100
Plutonium concentration (%)	15-35
O/M ratio (-)	1.94-2.00
Fractional porosity (%)	0-40
Grain size (μm)	3-30

During overpower transients high creep rates are seen especially if restructuring has occurred and fuel temperature is higher than 1800 K [7]. In agreement with Tab. 1, brittle fracture of oxide fuel occurs for values of stress ranging from 77 MPa up to 103 MPa in the temperature domain 1500 K - 2900 K.

3 CORRELATIONS IN THE OPEN LITERATURE

Correlations of thermal creep for MOX published in the open literature are briefly presented in following sub-sections. Correlations shown here are based on the hypothesis that creep is a superposition of two phenomena: a diffusional process that depends linearly on the stress and a dislocation process characterized by higher values of stress exponent and occurring at high values of temperature and stress.

3.1 Evans et al. (1971)

Authors have developed the correlation presented in Eq. 1 [8]. In this correlation the thermal creep is expressed in h^{-1}. Coefficients A and B are presented in Eq. 2 and Eq. 3. Meaning and domain of each parameter are following: σ is the stress (1000-10000 psi); T is the temperature (1473-1898 K); D is the fractional l density (0.88-0.95); G the grain size (4-35 μm); X is the fractional concentration of plutonium (0.2-0.3). R is the universal constant of gas (1.987 cal/K mol).

$$\dot{\varepsilon}_s = A\sigma \cdot exp\left(-\frac{100000}{RT}\right) + B\sigma^{4.5} \cdot exp\left(-\frac{140000}{RT}\right) \quad (1)$$

$$A = 3.1\ 10^{7} exp[33.3(1-D)]\left[\frac{exp(3.56X)}{G^3}\right] \quad (2)$$

$$B = 4.37\ 10^{-4} exp[10.3(1-D)]\ exp(3.56X) \quad (3)$$

3.2 Slagle et al. (1984)

Slagle and his co-authors have developed the correlation presented in Eq. 4 [9]. If compared to Eq. 1, this expression is composed of an additional term to account for the high-temperature creep noted in their experimental measurements. This high-temperature

contribution is based on measurements that have been performed in the temperature interval 2448 K - 2913 K for compressive stress ranging from 2.2 MPa to 6.5 MPa. In this correlation the thermal creep rate is expressed in s^{-1}. Coefficients A, B, C, and F are presented in Eq. 5-8. Meaning and unit of measure of each parameter are following: σ is the stress (MPa); T is the temperature (K); ρ is the fractional theoretical density (%); d the grain size (μm); R in the universal constant of gas (1.987 cal/K mol). Activation energies Q_1, Q_2, Q_3 are 92500, 136800, 300000 cal/mol, respectively. The low-temperature terms are consistent with the correlations presented in [7,10].

$$\dot{\varepsilon} = F\left\{A \cdot \sigma \cdot exp\left(-\frac{Q_1}{RT}\right) + B \cdot \sigma^{4.4} \cdot exp\left(-\frac{Q_2}{RT}\right) + C \cdot \sigma^{4.4} \cdot exp\left(-\frac{Q_3}{RT}\right)\right\} \quad (4)$$

$$A = \left(8.97\ 10^5 / d^2\right)[1 + 2.11(97 - \rho)] \quad (5)$$

$$B = \left(9.00\ 10^2\right)[1 + 0.22(97 - \rho)] \quad (6)$$

$$C = \left(2.87\ 10^{17}\right)[1 + 0.22(97 - \rho)] \quad (7)$$

$$F = exp[-43.8(1.97 - O/M)] \quad (8)$$

3.3 Dumbill et al. (1987)

Authors have developed a correlation of creep based on a review of experimental data; see Eq. 9 [11]. The thermal creep rate is expressed in s^{-1}. Factors indicated in Eq. 9 are presented in Eq. 10-12. Meaning and unit of measure of each parameter are following: σ is the stress (MPa); T is the temperature (K); p is the pore volume fraction (-); d_g is the grain size (m); Pu is the concentration of plutonium (wt. fraction of PuO_2); x is the deviation from stoichiometry that is expressed as $(U,Pu)O_{2+x}$. Eq. 9 has been derived imposing that primary creep rate is zero in the original form of correlation proposed by Dumbill et al. [11].

$$\dot{\varepsilon} = \left[f_1(p)\frac{\sigma}{d_g^2} + f_2(p)\ 4.8\ 10^{10}\sigma^{4.5} exp\left(-\frac{27000}{T}\right)\right] F(O/M, T) G(Pu) \quad (9)$$

$$G(Pu) = 0.2 + (16/3)Pu;\ if\ \ Pu > 0.15\ G(Pu) = 1.0 \quad (10)$$

$$f_1(p) = exp(3p);\ \ f_2(p) = exp(2.2p) \quad (11)$$

$$F(O/M, T) = 1.07\ 10^{-9} exp(-21000/T)\left\{[x^2 + 2000exp(-27000/T)]^{\frac{1}{2}} + x\right\}^2 + 4.03\ 10^{-7}|x|\exp(-40000/T) \quad (12)$$

3.4 European catalogue (1990)

In 1990 a group of experts from various European research organizations had gathered to edit a European catalogue of properties for $(U,Pu)O_2$ mixed oxide fuel [12]. In Eq. 13 one can see the correlation recommended for thermal creep. The original equation accounts for fission-induced creep through F that is the local fission. Our discussion has not considered this effect (F=0). The creep rate is expressed in s^{-1}. Meaning of symbols as above are: σ is the stress (MPa); p is the pore volume fraction (-); d the grain size (m).

$$\dot{\varepsilon} = f(p)\left\{\frac{\left(1.11 \cdot 10^{-6}\right)}{d^2} \cdot \sigma \cdot exp\left(-\frac{48000}{T}\right) + 5.20 \cdot 10^4 \cdot \sigma^{4.5} \cdot exp\left(-\frac{75000}{T}\right)\right\} \quad (23)$$

$$f(p) = exp(25p) \quad (34)$$

3.5 Malygin et al. (2010)

Most recent correlation considered in our analysis has been published by Malygin and his co-authors in 2010: see Eq. 15 [13]. This correlation is based on a statistical analysis of available experimental data and provides an explicit description of all the quantities mentioned in the introduction. Coefficients A and B account for the effect of porosity and plutonium concentration; see Eq. 16 and Eq. 17. Function R calculates the parameter employed in the rightmost factor of Eq. 15. R depends on the deviation from stoichiometry, temperature and activation energy ΔH_{fO}.

$$\dot{\varepsilon} = \left[\frac{A(P,\,C)\sigma}{d^2} + B(P,\,C)\sigma^{4.5} exp\left(-\frac{Q_1}{kT}\right)\right]\left[\frac{165}{R} exp\left(-\frac{Q_2}{kT}\right) + \frac{R}{1+450x^2} exp\left(-\frac{Q_3}{kT}\right)\right] \quad (45)$$

$$A(P,\,C) = Aexp(19P + 3.6C) \quad (56)$$

$$B(P,\,C) = Bexp(10.5P + 3.6C) \quad (67)$$

$$R = \left(\sqrt{x^2 + 82\exp(-\Delta H_{fO}/kT)} - x\right)^2 \quad (78)$$

This correlation expresses the thermal creep in h^{-1}. Coefficients A of Eq. 16 and B of Eq. 17 are $3.1\ 10^{+10}$ and $1.2\ 10^{+07}$, respectively. Meaning and domain of each parameter are following: σ is the stress (0.7-120 MPa); T is the temperature (1373-1948 K); P is the fractional porosity (0.03-0.10); d the grain size (3-25 µm); C is the fractional plutonium concentration (0.048-0.25). R is the universal constant of gas (1.987 cal/mol); x is the deviation from stoichiometry $(O/M - 2)$ with domain of validity 0.00-0.09. Activation energies Q_1, Q_2, Q_3, and ΔH_{fO} are $1.6\ eV$, $6.9\ eV$, $3.8\ eV$, $2.5\ eV$.

4 COMPARISON OF MODELS

Predictions of thermal creep models are compared to verify relevance and modelling of each input parameter. A reference set of input data has been adopted. This set is consistent with the experimental conditions in [10]. Thermal creep rate predictions have been calculated under this reference condition. Values of reference input variables are listed in Tab. 2. Afterwards, each input quantity has been in turn modified and creep rate re-calculated keeping remaining input variables at their initial values. Models' predictions are presented as a function of temperature in two distinct domains: from 1400 K up to 1900 K and from 1900 K up to 2900 K. Most models are assessed in the lower temperature domain. Slagle and his co-authors have performed experimental measurements in the temperature domain ranging from 2448 K up to 2913 K with stress up to 6.5 MPa. The high-temperature term of their model is based on this set of measurements; see Eq. 4. Concerning porosity, models are validated for values lower than presented in Tab. 1 that is up to around 12%.

Table 2: Reference input quantities

Input variable	Reference value
Stress (MPa)	20
Plutonium concentration (%)	25
O/M ratio (-)	1.97
Fractional porosity (%)	3
Grain size (µm)	20

4.1 Thermal creep in reference conditions

Predictions of models are in good agreement with some deviation at low temperature of Evans's model that showed to be a conservative option; see Fig 1.a. Slagle's model shows lowest increase of rate with temperature in the region below 2500 K whereas, the contribution of the high-temperature term becomes dominant in the second part of the temperature domain; see Fig. 1.b. Scatter of predictions has been evaluated by means of the average relative standard deviation of calculations (not considering Evans's ones). This quantity is about 36% in the low temperature domain and 59% in the high temperature domain up to 2400 K.

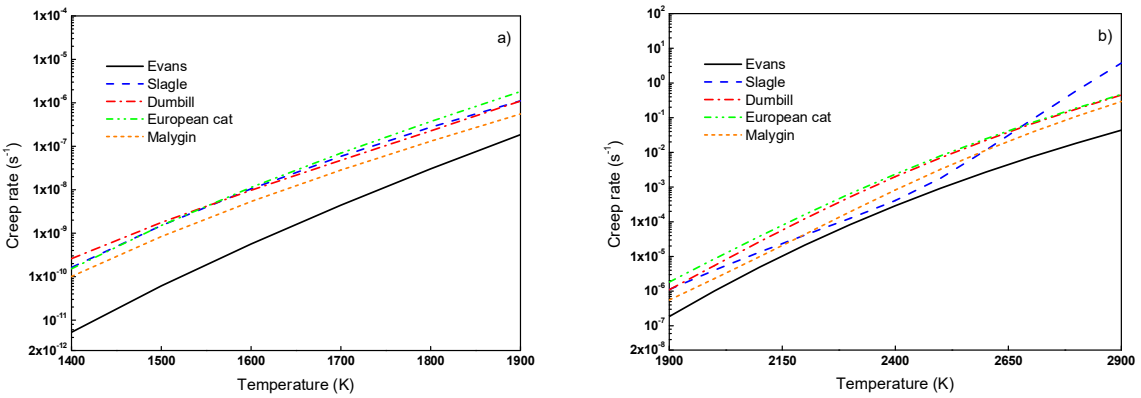

Figure 1: Creep rate for reference set of input data: low temperature a); high temperature b)

4.2 Effect of stress

This section presents a comparison of creep rate calculations by assuming an increase of stress from 20 MPa as reported in Tab. 2 to 60 MPa. This value induces a noticeable increase of creep rate, however, relative performance of models is rather consistent with the reference conditions. Compared to reference calculations, the increase moves from +722% (Slagle) up to +7177% (Evans) at 1800 K. Corresponding values increase to +7386% (Slagle) and +13164% (Evans) at 2400 K. This is also seen in the average relative standard deviation (as presented before) that increases to about 48% in the low temperature domain and to 84% in the high temperature domain up to 2400 K.

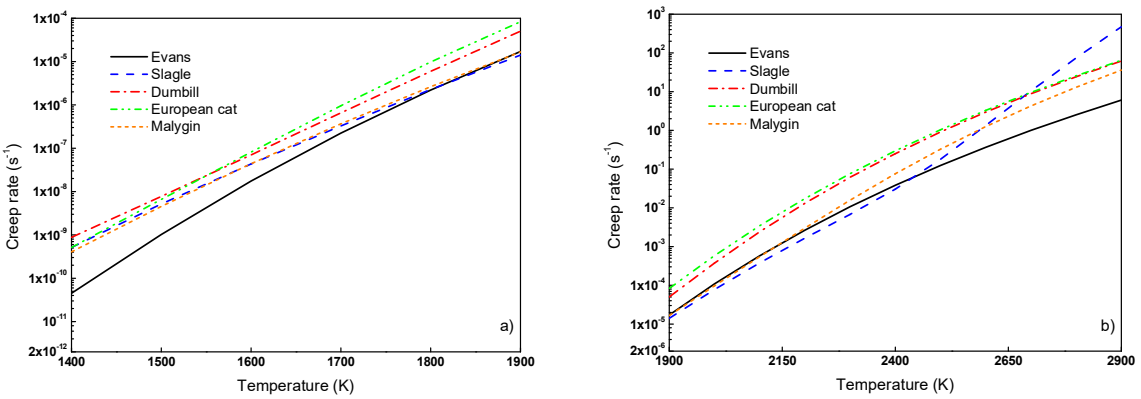

Figure 2: Creep rate by assuming a stress of 60 MPa: low temperature a); high temperature b)

4.3 Effect of porosity

Creep rate has been re-calculated by assuming an increase of porosity from 3% to 12%. This value induces an increase of creep rate accompanied with an increase of deviations between models. Relative increase is significant: +848% (European cat.), +1013% (Evans), +1827% (Slagle) at 1800 K. This observation is somehow confirmed in the high temperature domain. The relative increase predicted at 2400 K is +250%, +903%, +23%, +849%, +250% from Evans's to Malygin's model according to the legend shown in Fig. 3. Average relative standard deviations calculated as before move to 97% in the low temperature domain and to 103% in the high temperature domain (< 2400 K).

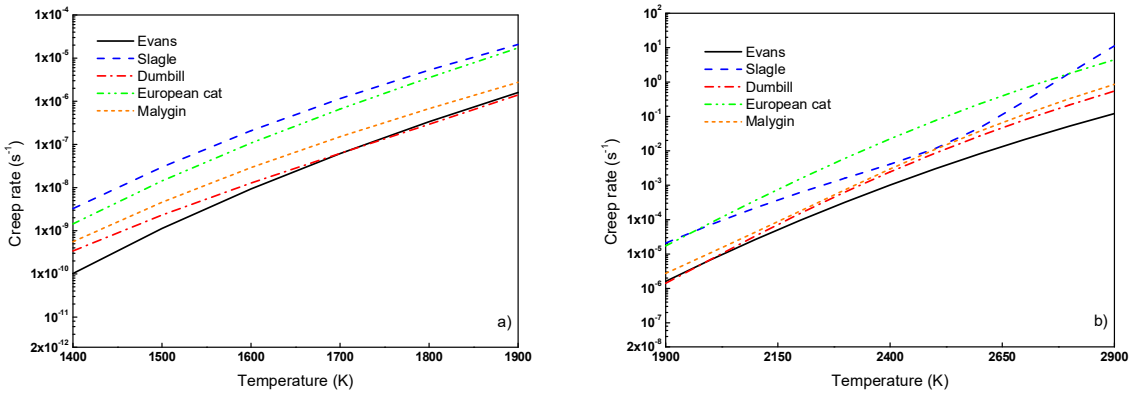

Figure 3: Creep rate by assuming a porosity of 12%: low temperature a); high temperature b)

4.4 Effect of plutonium concentration

This section presents creep rate calculations by assuming an increase of plutonium content from 25% to 30%. Only two correlations account for this modification: Evans and Malygin. Predictions of Dumbill's correlation are not modified in comparison with the reference case as plutonium concentrations are higher than 15%; see Eq. 10. A higher plutonium concentration leads to an increase by about +20% of the creep rate in comparison with reference values. Models showed to be in good agreement in predicting the effect of plutonium concentration; see Fig. 4.a and 4.b.

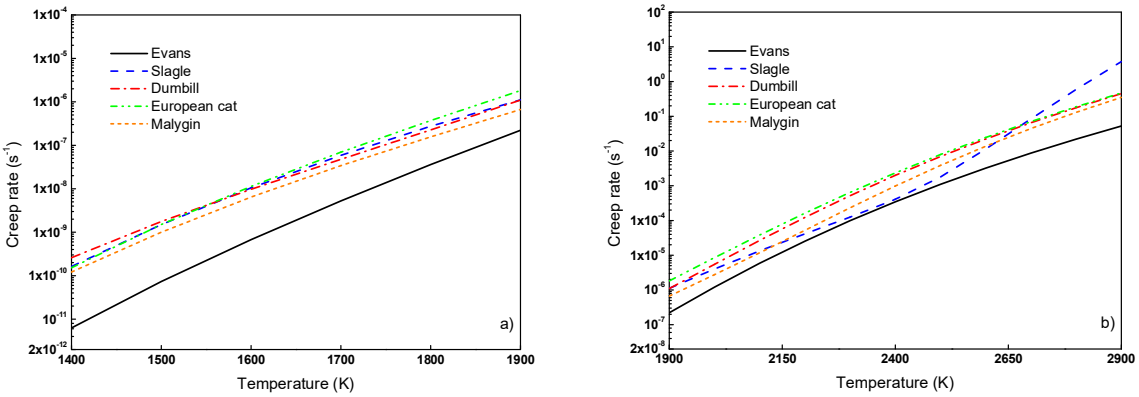

Figure 4: Creep rate by assuming a Pu content of 30%: low temperature a); high temperature b)

4.5 Effect of O/M ratio

This section presents a comparison between calculations by assuming a decrease of the O/M ratio from 1.97 to 1.90. The O/M ratio is not an input variable of Evan's and European catalogue models. In the low temperature domain Malygin's and Dumbill's model show a consistent increase of creep rate; see Fig. 5.a. At high temperature Malygin's model predicts a relative decrease while Dumbill is nearly unaffected. Slagle shows a relative decrease that is rather constant throughout both temperature domains; see Fig. 5.b. In the low temperature domain and in particular at 1800 K Dumbill's and Malygin's model agree on a relative increase around +170%. Slagle's model suggests a decrease by -95%. At 2400 K Slagle's, Dumbill's, and Malygin's models suggest a decrease by -95%, -1%, -40%, respectively. Average relative standard deviation moves to about 87% in the low temperature domain and to 81% in the high temperature domain (< 2400 K).

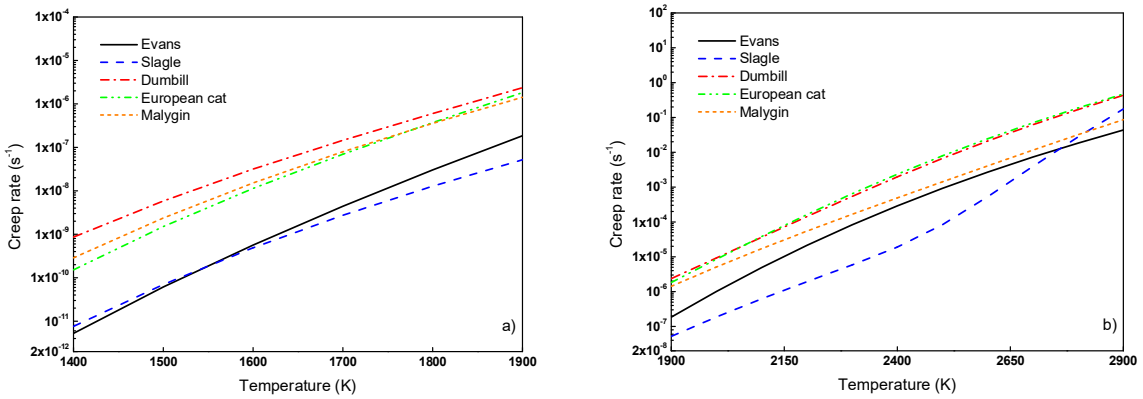

Figure 5: Creep rate by assuming a O/M ratio of 1.90: low temperature a); high temperature b)

4.6 Effect of grain size

This section presents a comparison between creep rates by assuming an increase of the gran size from 20 μm to 25 μm. This value of grain size causes a decrease of creep rate that is predicted in a rather consistent way by different models. Therefore, average relative standard deviations are about 37% in the low temperature domain and 68% in the high temperature domain again below 2400 K, both values being close to those calculated in reference conditions.

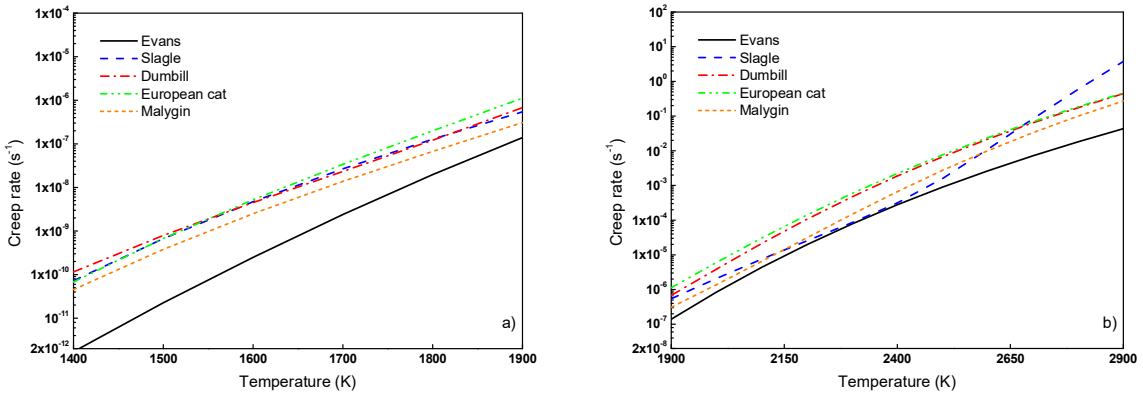

Figure 6: Creep rate by assuming a grain size of 25 μm: low temperature a); high temperature b)

5 CONCLUSIONS

This paper presents a comparison of thermal creep laws for FBR MOX. In general, correlations are in rather good agreement being Evans's model in most cases a conservative option. Changes of porosity and stoichiometry showed an increase in scatter of calculations. This could point out a need for more experimental data and an improved modelling of these parameters. This review has highlighted a need for new experimental data concerning: samples with higher values of porosity to meet FBR irradiation conditions; measurements conducted at high temperature and high stress conditions. Due to a lack in modelling, a deeper understanding of burn-up effect as done for MOX thermal conductivity turned out to be a relevant topic for thermal creep.

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