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Crystallographic Phase Transition of Zirconium Alloys: New Models for the TRANSURANUS Code

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ABSTRACT

The TRANSURANUS fuel performance code is featured by a clearly defined mechanical and mathematical structure which has permitted since its beginning a continuous development and an extension of the domains of application. In this view, the TRANSURANUS team has devoted significant efforts to make the code applicable for loss-of-coolant accident (LOCA) calculations. In parallel, besides standard Zircaloy-2 and Zircaloy-4, cladding material correlations for E110 that is used in VVER western-type reactor and, more recently, for the M5™ alloy of Framatome have been introduced in the code based on information in the open literature. In this view, the crystallographic phase transition of zirconium alloys having a strong impact on the high temperature creep and mechanical performance of the cladding is a topic relevant for LOCA simulations. The "*Reduction of Radiological Consequences of Design Basis and Design Extension Accidents*" project (R2CA) is partly funded by the European Commission within the H2020 programme. The objective of R2CA is to develop new calculation methodologies and updated computer codes in order to produce more realistic evaluations of radioactive releases resulting from LOCA or from Steam Generator Tube Rupture (SGTR) accidents. In the frame of R2CA (Task 3.2) ENEA, in coordination with JRC, has reviewed alternative models for the crystallographic phase transition and high-temperature creep of M5™. This activity aims at identifying new models for TRANSURANUS that are aligned with the state-of-the-art of research on these topics and, hopefully, capable of improving accuracy of code predictions during LOCA transients. Concerning the crystallographic phase transition of M5™, our review has identified the correlation published by Massih and Jernkvist. This recent model has been developed and validated with a complete and updated set of experimental data, however, it doesn't take into account the effect of hydrogen concentration on M5™. With regard to the high-temperature creep, a combination of Kaddour's model and, concerning the $(\alpha + \beta)$ region, a model by Massih, have been identified as an appropriate choice. These models have been implemented in the related TRANSURANUS subroutines and coupled as a standalone code for verification purposes. Some results that tackle main issues highlighted in our review are discussed throughout the paper. A specific part of the paper has been dedicated to the crystallographic phase transition of Zircaloy-4 with a proposal based on the model published by Massih in 2009. Verification will continue by introducing these models in the TRANSURANUS code to evaluate their performance in comparison with the existing models for LOCA calculations.

1 INTRODUCTION

TRANSURANUS, thanks to its clearly defined mechanical and mathematical structure, has been developed and extended aiming at proposing a versatile tool capable of tackling all

the conditions considered in licensing procedures [1,2]. In this view, significant efforts have been devoted to make the code applicable to LOCA transients. Noticeable advances have been achieved in the models employed for cladding ballooning [3,4] and cladding-steam reaction phenomena [5,6]. In parallel, code options for cladding materials have been extended by introducing a complete set of correlations for E110 which is employed in the Russian designed VVERs [5]. More recently, an identical action has been undertaken for the Zr-1%NbO (M5™) cladding material [7]. The crystallographic phase transition and high temperature creep play a relevant role for LOCA simulations. Code correlations of Zircaloy-4 have been updated to consider hydrogen concentrations up to 1000 ppm[1] and heating rates with absolute values up to 100 K/s [8,9].

In the frame of R2CA, ENEA, in coordination with JRC, has performed a review on M5™ correlations that has been presented in [10]. Results of this review have suggested, in agreement with the indications of the TRANSURANUS team, to consider in more detail the model published by Massih and Jernkvist (model B) for the crystallographic phase transition [11] and a combination of Kaddour's and Massih's models for the high temperature creep [12,13]. These models have been implemented in the TRANSURANUS subroutines for verification by means of a standalone program. Some results have been selected and presented in this paper. Additional considerations on Zircaloy-4 crystallographic phase transition have been included in the second part of the paper and a preliminary modelling proposed to the TRANSURANUS team. These models will be tested against current TRANSURANUS modelling of LOCA transients.

2 M5™ MODELS

2.1 Crystallographic phase transition

M5™ is a Zr-Nb alloy devoid of Sn with a controlled O, Fe and S content; see Tab. 1. M5™ is characterized by a homogeneous dispersion of β-Nb precipitates in fully recrystallized microstructure which enhances creep resistance [14]. The absence of Sn, the controlled alloy chemistry, and optimized heat treatment lead to a very high corrosion resistance and better mechanical properties in high burnup and high dose irradiations [14]. In the temperature domain of LOCA transients (600–1200 °C) crystals of zirconium-based alloys undergo a transition from a hexagonal close-packed (hcp) to a body centered-cubic (bcc) structure of the lattice. Phase transition is affected by hydrogen concentration and heat rate of thermal transients [8,15].

Table 1: Nominal composition of M5™ alloy (wt.%) [15]

	Sn	Nb	O	Fe	Cr	Ni	Zr
M5™	-	1.0	0.125	-	-	-	bal.

The model selected for the crystallographic transition of M5™ has been published by Massih and Jernkvist [10,11]. The β phase fractional volume y is calculated by means of Eq. 1.

$$\frac{\mathrm{dy}}{\mathrm{dt}} = \frac{1}{\tau(\mathrm{T})}\left(\left(\mathrm{y}_{eq}(\mathrm{T}) - \mathrm{y}\right) \pm \frac{\mathrm{b}}{\mathrm{y}_{eq}(\mathrm{T})}\left(\mathrm{y}_{eq}(\mathrm{T}) - \mathrm{y}\right)^2\right) \quad (1)$$

[1] The unit of measure *ppm* must be intended as *wt.ppm* throughout the paper.

Rate of β phase fractional volume is proportional to the deviation from equilibrium (y_{eq}) according to $\frac{1}{\tau(T)}$ where $\tau(T)$ in the characteristic time that is presented in Eq. 2.

$$\tau(T) = B \exp\left(\frac{E}{T}\right) \tag{2}$$

In Eq. 1 the plus sign is used for the description of heating transients while the minus sign is used for cooling transients. Authors introduced in this model an additional parameter b and a condition on the characteristic time $\tau(T)$: during cooling its value should not be higher than 6 s. This limit has been introduced to prevent that a transition from the β phase to the α phase remains incomplete [11]. The equilibrium phase transition of M5™ is shown in Eq. 3.

$$y_{eq} = \frac{1}{2}\left[1 + \tanh\left(\frac{T - T_{cen}}{T_{span}}\right)\right] \tag{3}$$

The fractional volume of β phase y_{eq} depends on T_{cen} and T_{span}. The first one is the middle temperature of the two-phase domain while the second one gives an indication on the width of this temperature domain; see Eq. 4.

$$T_{cen} = \frac{T_\alpha + T_\beta}{2}; \quad T_{span} = \frac{T_\beta - T_{cen}}{2.3} \tag{4}$$

Values proposed by authors for M5™ are: E = 54000 K, b = 0.3, $B = 1.55 \cdot 10^{-19}$ s. T_α and T_β are 1016 K and 1240 K, respectively [11]. This model doesn't consider the effect of hydrogen concentration. Based on the experimental findings, we assume that the effect of hydrogen on M5™ phase transition is consistent with the effect measured on Zircaloy-4 [8]. In our modelling the transition temperatures T_α and T_β are reduced as a consequence of the presence of hydrogen in compliance with the coefficients used for Zircaloy-4 [11]. Expressing the concentration of hydrogen in ppm, T_α is diminished by $1.52 \cdot 10^{-1}$ K per ppm and T_β by $2.20 \cdot 10^{-2}$ K per ppm. During transients the concentration of hydrogen affects the phase transition of Zircaloy-4 by means of a factor that modifies the characteristic time employed in Eq. 2 [11]. We have used this approach for M5™ employing a value of the constant in the exponential function that is higher than that used for Zircaloy-4 [11]; see Eq. 5. In the same way, coefficient B of Eq. 2 has been tuned to achieve phase transition curves that are consistent with the curves obtained for Zircaloy-4 at the same conditions of heating rate and hydrogen concentration [8].

$$\tau(T) = \frac{B \exp\left(\frac{E}{T}\right)}{1 - 0.0003\bar{x}_H\left(1 - \exp\left(\frac{5300}{T}\right)\right)} \tag{5}$$

Authors do not clearly state if this factor is to be applied during cooling transients [11]. Concerning Zircaloy-4, they do not present calculations on the effect of hydrogen during cooling where experimental measurements are lacking.

2.2 High temperature creep

The model proposed for M5™ is based on the correlation developed by Kaddour et al. that is presented in Eq. 6 [12].

$$\dot{\varepsilon} = \frac{A}{T}\sigma^n exp\left(-\frac{E}{RT}\right) \quad (6)$$

This correlation is applied in our proposal in the α and β single-phase domains as well as in the (α+β) two-phase domain for values of effective stress higher than 5 MPa. Experimental findings indicate that in the α phase, besides dislocation, a diffusion mechanism contributes to creep rate. The diffusion mechanism is dominant if values of stress are lower than 15 MPa, whereas above this threshold the creep strain rate is ruled by the dislocation mechanism. In the β phase the creep rate is described by a dislocation mechanism. Values of the pre-exponential constants, activation energies and stress exponents can be found in the original paper [12]. Concerning the (α+β) domain, a linear combination of single-phase creep rates is applied if values of stress are higher than 5 MPa; see Eq. 7.

$$\dot{\varepsilon}^{\alpha+\beta} = (1-y)\dot{\varepsilon}^{\alpha} + y\dot{\varepsilon}^{\beta} \quad (7)$$

Below this limit value, the creep rate is determined according to the model published by Massih [13]. This model is based on the theoretical approach to the superplasticity of zirconium alloys that was published by Ashby and Verrall [16]. Two mechanisms are used to describe the behaviour of M5™ creep rate in the two-phase domain: diffusion and dislocation. At high values of stress the diffusion mechanism is substituted by the dislocation one. In an intermediate region these two processes are superimposed and act independently (superplasticity). Correlations are given in Eq. 8 (diffusion term) and in Eq. 9 (dislocation term).

$$\dot{\varepsilon}_{diff}^{\alpha+\beta} = C_a\tilde{\sigma}\frac{\Omega\dot{\overline{D}}_V}{k_B T d^2}\left(1+\frac{\theta\delta\overline{D}_S}{d\overline{D}_V}\right) \quad (8)$$

In Eq. 8 C_a and θ are geometrical constants, Ω the atomic volume, d the grain size, $\overline{D}_V$ the average bulk diffusion coefficient, $\overline{D}_S$ the average grain boundary surface diffusion coefficient, δ the thickness of the grain boundary, and $\tilde{\sigma}$ is the effective stress.

$$\dot{\varepsilon}_{disl}^{\alpha+\beta} = \frac{1}{k_B T}\left(\frac{b}{d}\right)^p exp\left(-\frac{Q_d}{RT}\right)\left[A_\alpha\mu_\alpha\left(\frac{\sigma}{\mu_\alpha}\right)^{n_\alpha}(1-y) + A_\beta\mu_\beta\left(\frac{\sigma}{\mu_\beta}\right)^{n_\beta} y\right] \quad (9)$$

In Eq. 9 $A_{\alpha/\beta}$ is a material-dependent (dimensionless) constant in the α/β phase, $\mu_{\alpha/\beta}$ the shear modulus in the α/β phase, b the magnitude of the Burgers' vector, p an empirical inverse grain size exponent, and Q_d the activation energy for dislocation creep. Parameters of this model are p and $n_{\alpha/\beta}$. In these calculations the grain size remains constant with values lying in the interval 5-11 µm [13]. In compliance with this indication, the value of grain size is 6 µm. The original models show discontinuities at the boundaries between single-phase and two-phase domains. Concerning the concluding part of the (α+β) to β transition, Trego showed that a significant increase of grain size occurs in this region leading to a markedly decrease of creep in a narrow temperature interval [17]. Based on this indication, we have identified a small interval of the β fractional volume between the two- and single-phase region. In this domain the continuity of creep rate curves has been assured by construction with a linear combination of the values at the boundaries. A consistent approach has been used for the transition region from α to (α+β).

3 ZIRCALOY-4 MODEL

3.1 Crystallographic phase transition

Authors have reported that in the case of Zircaloy-4, model B shows deviations from the experimental measurements performed at high heating rates [11]. Aiming at discussing this inaccuracy of model B, we have developed phase transition correlations based on the first-order model proposed by Massih [18]. While most of the original model has been maintained unchanged, we have introduced an additional parameter. This factor is applied under the condition that the beta phase fractional volume is lower than 10%. The use of this factor, having values between 0.0 and 1.0, diminishes the phase transition rate. Above 10% of β phase fractional volume this factor is set to 1.0 so that the full phase transition rate is considered in calculations. This smooth increase of the transition rate is seen especially at high heating rates (+100 K/s) [19].

4 VERIFICATION OF THE MODELS

Phase transition and high temperature creep correlations presented in previous paragraphs have been introduced in the corresponding subroutines of TRANSURANUS. A standalone program that couples both subroutines has allowed us to perform calculations in the domains of interest concerning heating\cooling rate, hydrogen concentration, effective stress. Concerning the effect of hydrogen, an upper limit of 1000 ppm has been adopted. This value is consistent with the modelling of Zircaloy-4 conditions but much higher than observed at end of M5™ irradiations. Therefore, presented results are intended mainly for verification purposes of TRANSURANUS subroutines. Some results of our calculations are presented with the objective of considering issues of interest for future development or relevant for the following step of verification. In this second step we plan to perform LOCA calculations with TRANSURANUS. This will be a good opportunity to make a comparison between the predictions of models presented here with the results of existing modelling in TRANSURANUS.

4.1 Zircaloy-4 and M5™ crystallographic phase transition

Phase transition correlations (model B) of Zircaloy-4 and M5™ are compared at increasing values of heating\cooling rate neglecting the effect of hydrogen. Results are presented in Fig. 1. They confirm that the lower diffusion of niobium induces a delay of M5™ phase transition. Results at +10 K/s are quite consistent. A small delay of M5™ is noticed at +100 K/s [15].

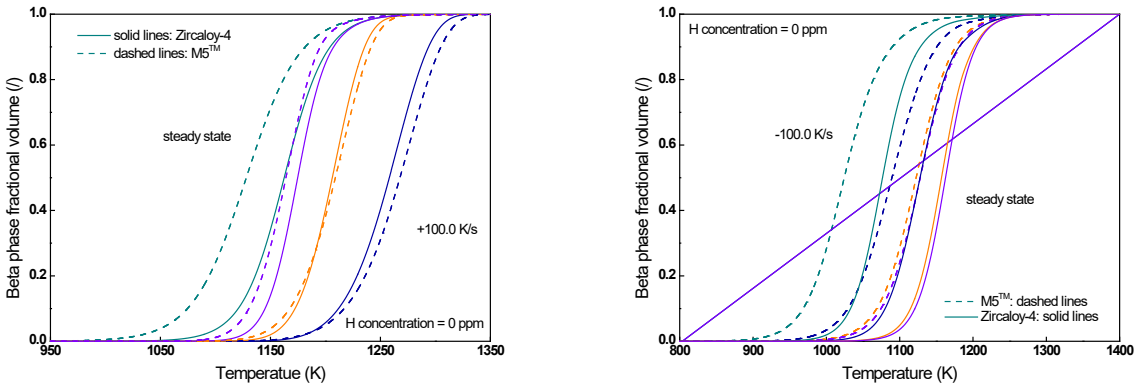

Figure 1: Zircaloy-4 and M5™ phase transition at 0, +1, +10, +100 K/s (left) and 0, -1, -10, -100 K/s (right)

Hydrogen pick-up is much more intense in Zircaloy4 than M5™. Figure 2 presents the phase transition curves of Zircaloy-4 if a concentration of 1000 ppm is assumed. According to the authors, the effect of hydrogen has been determined by using an expression that modifies the characteristic time as shown in Eq. 5 [11]. Results of Zircaloy-4 during cooling transients have been obtained taking into account only the effect of hydrogen on the transition temperatures that has been determined under quasi-equilibrium conditions. Due to the fast desorption of hydrogen at temperatures above the β transition temperature, it was not possible to perform the study of the inverse phase transformation kinetics upon cooling [8]. The use for Zircaloy-4 of the characteristic time as in Eq. 5 has shown that results obtained during cooling were loosely correlated with a possible effect of hydrogen (not shown). Based on these considerations, it has been decided to neglect this factor during cooling transients where, as above mentioned, the effect of hydrogen is not well established.

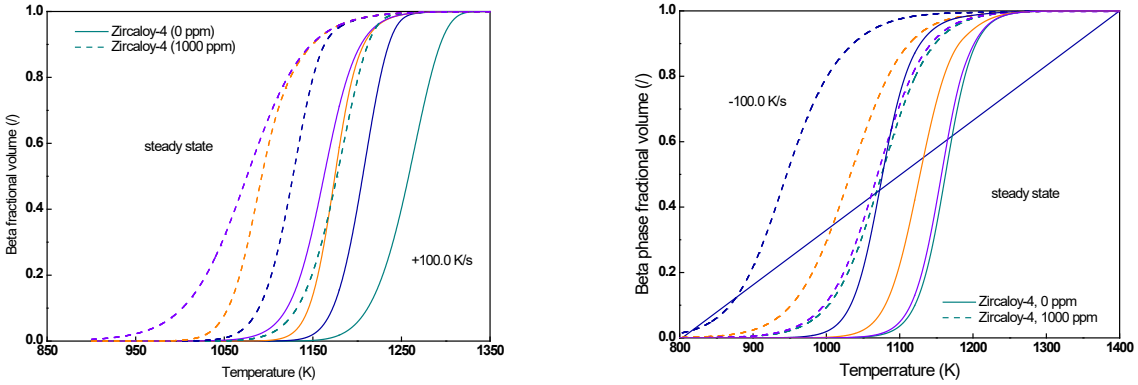

Figure 2: Zircaloy-4 phase transition at 0, +1, +10, +100 K/s (left) and 0, -1, -10, -100 K/s (right); effect of hydrogen concentration (1000 ppm)

4.2 Effect of hydrogen concentration on M5™ crystallographic phase transition

As shown in Fig. 2, the effect of hydrogen is of great importance for Zircaloy-4 phase transition. The use of M5™ is quite beneficial from this point of view as much lower concentrations of hydrogen are expected [14]. Nevertheless, the effect of hydrogen on M5™ is of interest to make the code capable of estimating its effect and of treating an ample domain

of conditions. The duty in charge of ENEA within Task 3.2 of R2CA is to provide an updated model/subroutine for M5™ that could cope with domains of applicability comparable with the existing TRANSURANUS model of Zircaloy-4 [9]. A comparison of the model of M5™ [11] with the modified one assuming increasing concentrations of hydrogen is presented in Fig. 3. The interval of concentrations considered in the comparison ranges from 0 ppm up to 500 ppm.

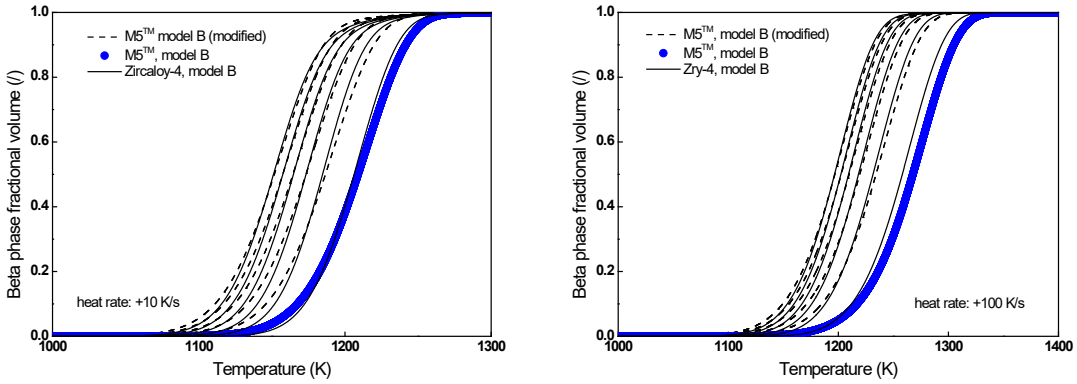

Figure 3: Phase transition with a concentration of hydrogen in the interval 0-500 ppm (every 100 ppm): comparison of models at +10 K/s (left) and +100 K/s (right) [11]

Values of hydrogen concentration representative of the operating conditions and burn-ups typical of M5™ irradiations lie in the interval 60-100 ppm. In agreement with the findings presented in [8], our results confirm that predictions of the model proposed for M5™ are quite consistent with Zircaloy-4. Same conclusion can be drawn by applying a heating rate of +100 K/s.

4.3 Creep rate of M5™ in the (α+β) region

Literature has underlined that modelling of M5™ creep rate is not well established in the (α+β) region [12,20]. Results of the model proposed here are presented in Fig. 4. In this figure the creep rate of M5™ has been calculated as a function of temperature under two heating rate conditions: +10 and +100 K/s. Values of stress employed in calculations are 3 MPa, 10 MPa, 20 MPa, and 30 MPa.

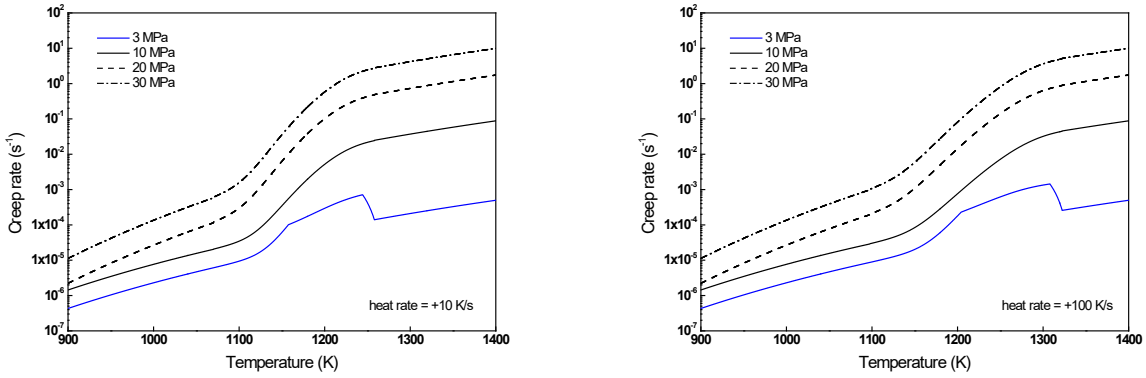

Figure 4: Creep rate of M5™ as a function of temperature and heat rate: +10 K/s (left) and +100 K/s (right)

The first value lies in the stress domain where it has been assumed that Massih's model is applied [13]. Results have been obtained by assuming a constant value of grain size (6 μm). Transition regions from Kaddour's to Massih's model and back from Massih's to Kaddours's model across the (α+β) region have been tentatively imposed. Under these conditions our results show a peak of creep rate at 3 MPa that tends to shift toward higher temperatures and to increase with increasing heating rate. Calculations are consistent with predictions presented in the literature.

4.4 Preliminary model for phase transition of Zircaloy-4

In this section we discuss a comparison of predictions obtained with two different models of Zircaloy-4 phase transition: model B [11] and model A [18]. Correlations of model A have been partly modified on purpose. As stated by authors, model B shows deviations from the experimental results of at high heating rates [11,19]. Results of models at +5, +10, +50, and +100 K/s are presented in Fig. 5. Model A shows reasonable agreement with model B at low values of heating rate and a more satisfactory agreement with the experimental results at +100 K/s [19]. While great part of model's correlations proposed in [18] has not been modified, a tuning factor of absolute value lower than 1.0 has been applied to the rate of phase transition when the β fractional volume is lower than 10%. This proposal relies on the hypothesis that the full rate of phase transition is attained in the region above 10% of beta fractional volume. This hypothesis has been confirmed especially at high heating rates in [19], moreover, uncertainties affecting beta volume measurements are higher below 10% [8,15]. In principle, this tuning factor could improve accuracy of phase transition predictions especially at high heating rates where model B tends to underestimate the experimental measurements [8,19].

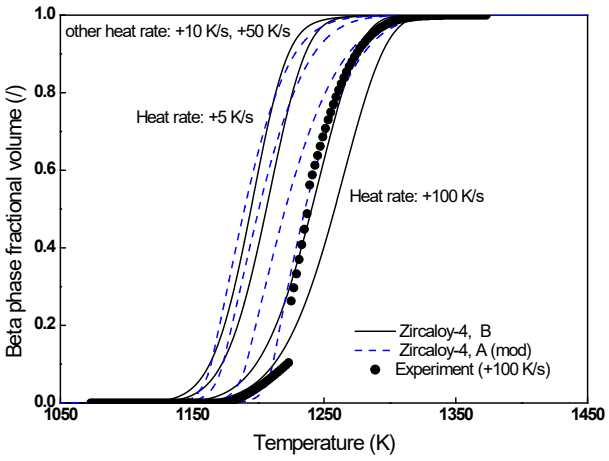

Figure 4: Phase transition of Zircaloy-4: comparison of model B and model A (mod.) with experimental measurements [19].

5 CONCLUSIONS

This paper presents some results on the activity performed by ENEA in coordination with JRC within Task 3.2 of R2CA. Our contribution is focused on the identification of correlations for the crystallographic phase transition and high-temperature creep of M5™ that could update and eventually improve the existing models of TRANSURANUS. The review

has identified some models fitting for our purpose. They have been presented and results of corresponding TRANSURANUS subroutines discussed in this paper. If on the one hand, it has been confirmed that predictions are consistent with the literature, on the other hand, relevant aspects are still not well established such as the effect of hydrogen during cooling transients and creep rate across the phase transition of M5™. In this view, our proposal tends to be conservative with regard to the effect of hydrogen (no modelling during cooling transients) while it has been attempted to model the creep rate in the two-phase domain. Besides this, preliminary indications on a novel phase transition model for Zircaloy-4 have been proposed in the concluding part of the paper. In principle, this model could improve accuracy of predictions especially at high heating rates where model B showed to be less precise. In conclusion, this stage of verification has given positive indications addressing several aspects of modelling relevant for TRANSURANUS. In the following step these models will be compared to the existing modelling by performing code calculations on LOCA experimental tests. It is expected that this future activity could provide recommendations useful to the community of TRANSURANUS users.

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