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Review of M5™ Cladding Models Relevant for LOCA Simulation with the TRANSURANUS Code

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ABSTRACT

Reliable and accurate fuel performance codes are crucial for the design of nuclear fuel. Codes should have the capability of predicting fuel behaviour under both normal operations and postulated accidents. Based on the fact that phenomena occurring under these two conditions are quite different, distinct codes have been historically developed for the purpose. The TRANSURANUS 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 its application.

In particular, significant efforts have been devoted by the developers to make the code applicable for loss-of-coolant accident (LOCA) calculations. In parallel, cladding material options have been extended. Besides standard Zircaloy-2 and Zircaloy-4, 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 the frame of the *Reduction of Radiological Consequences of design basis and design extension Accidents* project (R2CA) of EURATOM, ENEA, in coordination with the JRC, has reviewed alternative models for the crystallographic phase transition and high temperature creep of M5™. Both quantities are relevant for LOCA simulations. The final goal of this work is to identify correlations that could improve accuracy of code predictions. This analysis aims at providing an adequate source of information to achieve this objective.

1 INTRODUCTION

As mentioned in the abstract, 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 modelling of clad ballooning [3,4] and cladding-steam reaction [5,6]. In parallel, options for cladding materials have been extended with the E110 alloy which is employed in the Russian designed VVERs [5]. More recently, correlations for Zr-1%NbO (M5™) alloy have been introduced in the code [7]. In strict compliance with the developments of modelling, an intense activity for verification and validation under LOCA conditions was conducted especially in the frame of international codes' benchmarks [8, 9].

The crystallographic phase transition and high temperature creep are certainly relevant for LOCA simulations. Code correlations of Zircaloy-4 have been updated to consider a hydrogen concentration up to 1000 ppm and heating rates up to 100 °C/s [10,11]. In the frame

of R2CA, ENEA should propose and implement correlations for M5™ that could cope with domains of hydrogen concentration and non-isothermal conditions close to those achieved for Zircaloy-4. Models discussed for the purpose are presented in this paper.

2 CRYSTALLOGRAPHIC PHASE TRANSITION

In the temperature domain typical 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. This behaviour has been confirmed also in the case of the M5™ alloy [12]. Phase transition is affected by the concentration of hydrogen and the heat rate of transients. Therefore, a distinction is necessary between isothermal or equilibrium conditions and dynamic thermal conditions [10,12].

Table 1: Nominal composition of M5™ alloy (wt.%) [12]

	Sn	**Nb**	**O**	**Fe**	**Cr**	**Ni**	**Zr**
M5™	-	1.0	0.125	-	-	-	balance

The correlation implemented in the latest version of the code (v. 2021) is an analytical curve depending on temperature and thermal conditions that are either isothermal or non-isothermal [7]. This latter condition corresponds to the experimental curve obtained by applying a heating rate of +100 °C/s [12].

As done for the BISON code [13], our analysis has started from the model published by Massih [14]. A second, more recent model has been used for comparison [15]. Massih's model is presented in Eq. 1 and the model by Massih&Jernkvist in Eq. 2. The volume fraction of β phase (y) varies in proportion with its deviation form equilibrium (y_{eq}).

$$\frac{dy}{dt} = \frac{1}{\tau(T)}\left(y_{eq}(T) - y\right) \tag{1}$$

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

Rate and deviation from equilibrium are correlated through the $k(T)$ term that is presented in Eq. 3. This quantity is the reciprocal of the characteristic time $\tau(\mathrm{T})$. In Eq. 3 $\underline{k_B}$ is the Boltzmann constant, E the activation energy, and T is the temperature.

$$k(T) = \frac{1}{\tau(\Gamma)} = k_0 \exp\left(-\frac{E}{k_B T(t)}\right) \tag{3}$$

In Eq. 2 the plus sign is used for the description of heating transients while the minus sign is used for cooling transients. This model does not employ the transition temperatures T_α and T_β. These quantities represent the lower and the upper bound of the temperature domain across which phase transition occurs. The authors introduced in this model an additional parameter b and a condition on the characteristic time $\tau(\mathrm{T})$: during cooling its value should not be higher than 6 s. This choice prevents from an interruption of phase transition that would lead to a final value higher than zero [15]. The equilibrium phase transition curve (y_{eq}) of M5™ is expressed by means of Eq. 4.

$$\mathrm{y}_{eq} = \frac{1}{2}\left[1 + \tanh\left(\frac{\mathrm{T} - \mathrm{T}_{cen}}{\mathrm{T}_{span}}\right)\right] \tag{4}$$

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 temperature domain while the second one gives an evaluation of the width of this domain; see Eq. 5.

$$T_{cen} = \frac{T_\alpha + T_\beta}{2}; \quad T_{span} = \frac{T_\beta - T_{cen}}{2.3} \tag{5}$$

Our proposal is to consider, as already done for BISON [13], the first-order model as reference (Eq. 1). The equilibrium curve and the behaviour during cooling should be defined for the purpose stated that, in the case of M5™, the author has not included this information in his work due to a lack of experimental data [14]. Based on the values of T_α and T_β reported in [15] the equilibrium curve of M5™ has been determined according to Eq. 5. As shown in Fig. 1 (left), this curve proved to be in quite good agreement with the corresponding curve given by Forgeron et al. that was expressed by means of a Johnson-Mehl-Avrami-Kolmogorov correlation [12]. The effect of hydrogen concentration on the equilibrium curve has been determined by applying a similar approach. The analytical correlations based on the experimental measurements conducted on Zircaloy-4 [10] have been compared with the model of Zircaloy-4 reported in [15]. The curve used in this work is presented in Fig. 1 (right) together with just mentioned curves. As one can note in this figure, the curve used in calculations is in slight better agreement with the curve presented in [10] thanks to an increase in the width of the temperature domain.

The level of hydrogen concentration expected for M5™ is 5 to 6 times lower than Zircaloy-4, nevertheless it is assumed that, concerning the effects due to the presence of hydrogen, both alloys do not differ significantly [10]. A concentration of 100 ppm has been considered well representative of the operating conditions of M5™.

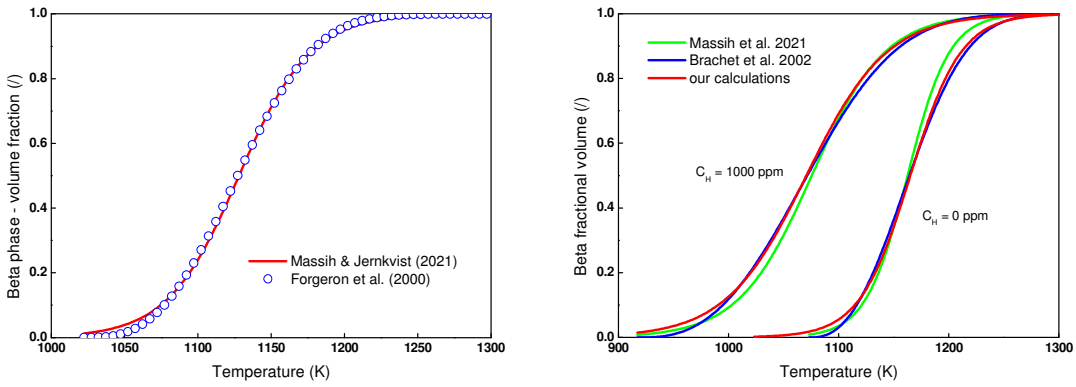

Figure 1: M5™ – equilibrium curve (left); effect of hydrogen concentration on Zircaloy-4 (right)

The analysis of cooling transients of Zircaloy-4 is performed in BISON by introducing in Eq. 3 an additional constant. This constant accounts for a martensitic contribution to phase transition [13]. As aforementioned, the model discussed in [15] adopts specific indications during cooling: use of the minus sign and an upper bound of the characteristic time τ(T). The approach applied during cooling transients in our model is to use a virtual equilibrium curve shifted towards lower temperatures. The absolute value of the shift is increasing with increasing cooling rates. The characteristic time has been determined to comply with the indications provided by the model presented in [15]. In the case of M5™, this latter model does not depend on hydrogen concentration [15]. Therefore, an effect consistent with that adopted for Zircaloy-4 [15] has been introduced for comparison. Tables 2 and 3 report the

details of the model proposed here. In these tables w is the concentration of hydrogen in ppm and Q the heat rate in °C/s.

Table 2: Model proposed for heating transients

Heating		
Heat rate conditions	$0 < Q\ (°C/s) \leq 0.1$	$0.1 < Q\ (°C/s) \leq 100$
$\mathbf{T_{start\_\alpha}}$ (K)	$1060.0 - 0.156w$	$[(1118.0 - 0.156w)Q^{0.023}]$
$\mathbf{K_0}$ (s^{-1}); $\mathbf{E/K_B}$ (K)	$60457 + 18129Q$; 16650	
$\mathbf{T_{cen}}$ (K)	$1128.0 - 0.094w$	
$\mathbf{T_{span}}$ (K)	$48.70 + 0.027w$	

Table 3: Model proposed for cooling transients

Cooling		
Heat rate conditions	$-0.1 \leq Q\ (°C/s) < 0.0$	$-100 \leq Q\ (°C/s) < -0.1$
$\mathbf{T_{start\_B}}$ (K)	$1240.0 - 0.032w$	$1240.0 - 0.032w$
$\mathbf{K_0}$ (s^{-1}); $\mathbf{E/K_B}$ (K)	$2.6 \cdot 10^{+07}[ln(\lvert Q\rvert + 0.9)]^{1.5}$; 16650	
$\mathbf{T_{cen}}$ (K)	$1128.0 - 0.094w - 20\ln(\lvert Q\rvert + 0.9)$	
$\mathbf{T_{span}}$ (K)	$48.70 + 0.027w$	

Fig. 2 suggests good agreement between models under the hypothesis that the concentration of hydrogen is negligible. At higher concentration (100 ppm) some deviation has been noted. These results are not presented here as they need further verification.

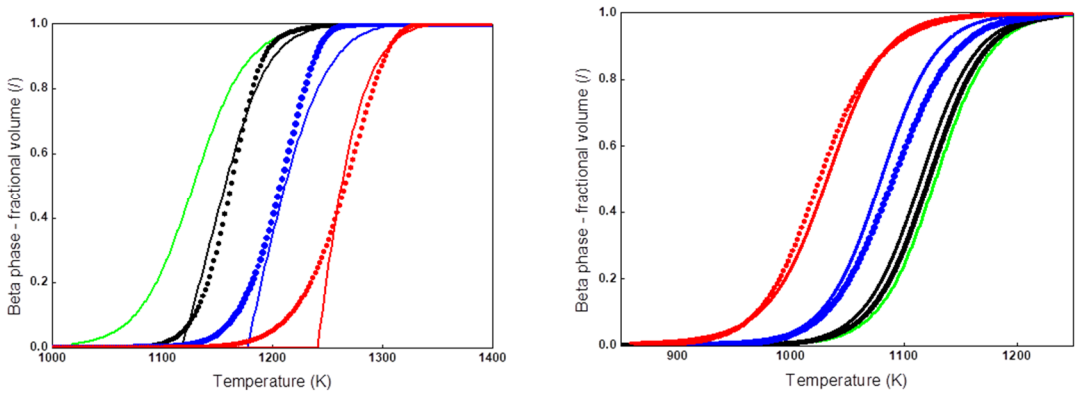

Figure 2: Comparison of proposed model (solid lines) with predictions of [15] (scatter) - curves during heating (left) and cooling transients (right) calculated at a rate of ±1 (black), ±10 (blue), ±100 (red) °C/s. In this figure the equilibrium curve is represented by the green solid line.

3 HIGH TEMPERATURE CREEP

In TRANSURANUS (v. 2021) the high temperature creep of M5™ is based on the corresponding correlation of Zircaloy-4 [7]. This correlation is a Norton law which is determined by three parameters: the structure constant A, the activation energy E, and n that is the stress exponent. In addition, a stress correction factor has been introduced to scale model's

predictions to M5™ alloy [7]. In this section we discuss three high temperature creep models for M5™. The first one has been developed by Kaddour et al. and it is presented in Eq. 6 [16].

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

The experimental findings indicated that in the α phase, besides dislocation, a diffusion mechanism contributes to creep rate. Below a threshold value of stress (σ_{th} =15 MPa) the diffusion mechanism is dominant, whereas above this threshold the creep strain rate is determined by a dislocation mechanism. Concerning the (α+β) domain, the authors could not define a simple correlation. In this region the experimental results proved to be higher than foreseen by a combination of creep correlations valid in the α and β domain.

The second model was proposed by Trego [17]. His experimental results confirmed both the co-existence of diffusional and dislocation mechanisms in the α domain and a pure dislocation behaviour in the β domain. The author has tackled the open issues highlighted by Kaddour et al. concerning the behaviour of M5™ creep in the (α+β) domain [16]. A series of experimental investigations focused on the two-phase domain and especially on the so-called quasi-β domain were conducted [17]. The quasi-β domain is a narrow temperature region where the fractional volume of the β phase moves from 92% up to about 98%. A significant increase of the β grain size was measured in this region. The author proposed, based on the indications of the literature, to include the effect of grain size in the modelling of M5™ creep rate.

Trego introduced a diffusional regime of creep strain rate in the quasi-β and β domain. A threshold value of stress fixes the transition from a diffusion mechanism to a dislocation one. The diffusion term in the α phase domain is presented in Eq. 7. The dislocation term has remained unchanged and in this region it is expressed by means of Eq. 6. The diffusional and dislocation correlations suggested by Trego for the quasi-β and β domain are reported in Eq. 8 [17].

$$\dot{\varepsilon}_{diff} = \frac{A}{T}\sigma^{n_1}\left(\frac{d_0}{d_\alpha}\right)^{3} exp\left(-\frac{E}{RT}\right) \tag{7}$$

$$\dot{\varepsilon}_{diff} = \frac{A}{T}\sigma^{n_1}\left(\frac{d_1}{d_\beta}\right)^{3} exp\left(-\frac{E}{RT}\right); \quad \dot{\varepsilon}_{disl} = \frac{A}{T}\sigma^{n_2}\left(\frac{d_\beta}{d_2}\right)^{2} exp\left(-\frac{E}{RT}\right) \tag{8}$$

As one van note in Eq. 7 and 8, this model employs relative values of grain size. The first parameter d_0 represents a reference value of the initial α grains size. The second one d_1 is the size of β grains at the onset of the quasi-β domain. The last one d_2 is a threshold value above which the dislocation term is unaffected by the β grains size. Values adopted in calculations are 7, 20 and 90 μm, respectively. The β grain size increases across the quasi-β domain from 20 up to 70-100 μm. In the 100% β domain this quantity may reach hundreds of microns.

The third model has been published by Massih in 2013 [18]. This model employs a theoretical approach to the so-called superplasticity behaviour of zirconium alloys that was presented by Ashby and Verrall in [19]. Two mechanisms are used to describe the behaviour of M5™ creep rate in the two-phase domain: diffusional and dislocation. Besides a standard representation of the dislocation term, the author extends the applicability of the diffusion model by Ashby and Verrall to the two-phase domain [18]. The diffusion mechanism is substituted by the dislocation one at high values of stress. In an intermediate region these two

processes are superimposed and act independently. This condition is called superplasticity. Correlations are presented in Eq. 9 (diffusion term) and in Eq. 10 (dislocation term).

$$\dot{\varepsilon}_{diff}^{\alpha+\beta} = C_a \tilde{\sigma} \frac{\Omega \dot{\bar{D}}_V}{k_B T d^2} \left(1 + \frac{\theta \delta \bar{D}_S}{d \bar{D}_V}\right) \tag{9}$$

In Eq. 9 Ca and θ are geometrical constants, Ω the atomic volume, d the grain size, $\bar{D}_V$ the average bulk diffusion coefficient, $\bar{D}_S$ the average grain boundary surface diffusion coefficient, δ the thickness of the grain boundary, and $\tilde{\sigma}$ is an effective stress.

$$\dot{\varepsilon}_{disl}^{\alpha+\beta} = \frac{1}{k_B T} \left(\frac{b}{d}\right)^p exp\left(-\frac{Q_d}{RT}\right) \left[A_\alpha \mu_\alpha \left(\frac{\sigma}{\mu_\alpha}\right)^{n_\alpha} (1-y) + A_\beta \mu_\beta \left(\frac{\sigma}{\mu_\beta}\right)^{n_\beta} y\right] \tag{10}$$

In Eq. 10 $A_{\alpha/\beta}$ is a material-dependent (dimensionless) constant in the α/β phase, $\mu_{\alpha/\beta}$ the shear strain 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 $n_{\alpha/\beta}$. The author in his analysis has applied values of grain size lying in the interval 5-11 µm. This quantity remains constant during phase transition [18].

This section aims at discussing predictions of these models. Some assumptions have been made for the purpose. The first one concerns the grains size. The curve of β grains size used in our calculations is presented with the experimental values [17] in Fig. 3 (left). The grains size of the α phase is constant (9 µm). In the model by Massih (Eq. 9 and 10) it has been assumed that this quantity is the sum of α and β grain size weighted by the fractional volume of β phase. A second assumption deals with the evaluation of creep rate in the two-phase domain. This quantity has been calculated according to Eq. 11.

$$\dot{\varepsilon}^{\alpha+\beta} = (1-y)\dot{\varepsilon}^{\alpha} + y\dot{\varepsilon}^{\beta} \tag{11}$$

This approach has been used for the model by Kaddour et al. and the model by Trego. In this latter case Eq. 11 has been applied in the region where the fractional volume of the β phase moves from 10% (end of quasi- α domain) up to 92% (start of quasi-β domain).

Calculations are based on additional assumptions as follows:

- equilibrium curve of M5™ based on [12];

Trego (2011)

- d_0 = 7 µm, d_1 = 20 µm, d_2 = 90 µm;
- end of quasi-α = 1077.15 K (y = 10%);
- start of quasi-β = 1186.15 K (y = 92%);
- end of quasi-β = 2010.15 K (y = 97.7%);

Massih (2013)

- $A_\alpha = A_\beta = 0.0010$;
- $n_\alpha = n_\beta = 3.50$.

For the sake of brevity, remaining parameters of Massih's model have not been reported but details are provided in [18]. For the same reason, the reader can find the coefficients of Kaddour et al. and Trego's model in the original papers [16,17]. Results in the low stress domain (1-5 MPa) are presented in Fig. 3 (right). Bottom and uppermost curves have been

calculated by assuming a value of stress if 1MPa and 5 MPa, respectively. An increase by 1 MPa is assumed moving from one curve to the following. Experimental tests conducted in this region are presented in [16]. Predictions at higher values of stress (25-60 MPa) are presented in Fig. 4 (left). Bottom and uppermost curves have been calculated by assuming a value of 25 MPa and 60 MPa, respectively. An increase by 5 MPa is assumed moving from one curve to the following. Finally, a comparison between Massih's and Kaddour's models limiting to the two-phase domain is presented in Fig. 4 (right). These results confirm that the use of the experimental values of grain size and their inclusion in the modelling improve the accuracy in predicting the behaviour of M5™ creep rate in the (α+β) domain. Models show a satisfactory agreement in the single-phase domains while in the two-phase domain the indications of Kaddour's and Trego's models show deviations both in the low and high stress domains. We recall a lack of experimental data in the high stress domain.

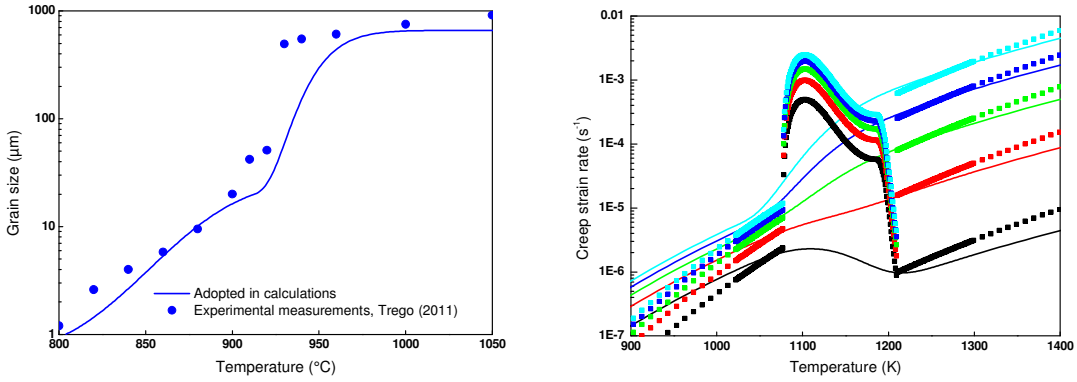

Figure 3: Experimental data and curve of β grain size as a function of temperature (left); M5™ high temperature creep (1-5 MPa): comparison of Kaddour's (solid) with Trego's model predictions (scatter) (right)

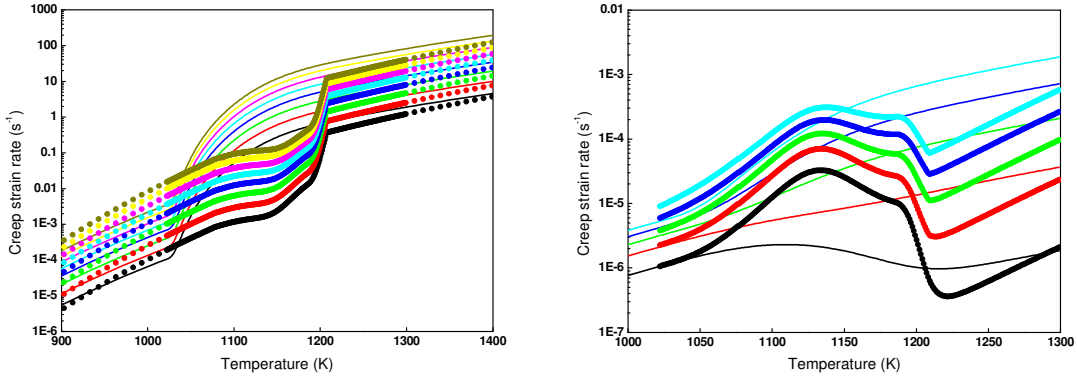

Figure 4: M5™ high temperature creep (25-60 MPa): comparison of Kaddour's (solid) and Trego's model predictions (scatter) (left); M5™ high temperature creep (1-5 MPa): comparison of Kaddour's (solid) and Massih's model predictions (scatter) in the two-phase domain

4 CONCLUSIONS

In the frame of R2CA of EURATOM, ENEA has reviewed two properties of the M5™ alloy that are relevant for simulation of LOCA transients: the crystallographic phase transition and the high temperature creep. This activity was performed in coordination with the JRC and aimed at identifying improvements and refinements of the modelling implemented in the

standard version of the TRANSURANUS code. A model for M5™ phase transition based on the work published by Massih in 2009 has been proposed. This model maintains the indications provided in the original paper concerning heating thermal conditions and introduces a modelling of cooling transients and hydrogen concentration.

Models of high temperature creep reviewed in this paper have confirmed the indications of the literature and the role played by the increase of the β grain size in the (α+β) domain. A lack of experimental data is a major limitation for final conclusions. However, these results are expected to be of help to identify new correlations for M5™ that could cope with available indications of the literature, on the one hand, and code requirements, on the other hand.

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