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Validation of the TRANSURANUS code: Melting temperature of MOX fuel for fast reactors

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Sommario

Nel documento si analizzano varie correlazioni per il calcolo della temperatura di fusione del combustibile MOX per reattori veloci. A questo scopo, dopo aver raccolto i dati sperimentali pubblicati, è stata valutata la capacità predittiva dei modelli disponibili nel codice TRANSURANUS e in letteratura. L'analisi critica dei risultati ha consentito di ricavare indicazioni utili allo sviluppo del codice.

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Note

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CONTENTS

1 INTRODUCTION

The fuel melting temperature is one of the key properties for the design of nuclear systems and the assessment of their safety performance under normal and accident conditions [1]. Melting temperature, usually intended as solidus temperature, decreases under irradiation due to the build–up of fission products in the fuel matrix. Other parameters are of great importance in the evaluations of this physical property, markedly: plutonium concentration, O/M ratio and the concentration of minor actinides (MAs). In MOX fuel for fast breeder reactors (FBRs) the high content of plutonium plays a prominent role. The report presents a review of the experimental findings and, based on this information, an assessment of the status of TRANSURANUS. In order to improve the code performance, the accuracy of other models published in literature was investigated to identify promising correlations.

In the following sections, the options offered by the code and the models discussed in the document are briefly described. Afterwards, the mentioned review and the results of the assessment are presented. In the final section, some conclusions are given.

2 TRANSURANUS MODELLING

The code is capable of simulating phase changes [2]. In addition to the thermophysical properties, the solidus temperature is one of the input parameters of important models such as the formation/closure of central void and the thermal strain between solid and liquid phases [2]. For FBR applications, the code offers two correlations given in equations (1) and (2). While in the former a conservative approach is adopted, accounting for the scatter of experimental data with a shift of 40 K, the latter is the correlation recommended for the analysis of fast reactors.

$$T_{m,sol} = 2800 - 450 c_{Pu} - 60 c_{Pu} \cdot (1 - c_{Pu}) + 273.15 - \Delta T_{O/M} - \Delta T_{bu} \tag{1}$$

$$T_{m,sol} = 2840 - 450 c_{Pu} + 273.15 - \Delta T_{O/M} - \Delta T_{bu} \tag{2}$$

These relationships depend on the local concentration of plutonium (c_{Pu}) taking into account the impact of the O/M ratio and burn–up according to the following two terms:

$$\Delta T_{O/M} = 10^4 \cdot (O/M - 2)^2 \tag{3}$$

$$\Delta T_{bu} = 7bu \tag{4}$$

In (4), the value of burn–up (*bu*) is expressed in at.%.

In regard to the effect due to the presence of plutonium, the code predicts the decrease of melting point by a value of about 400 K (for a concentration of 100 wt.%

– pure PuO_2); see Fig. 1. In the domain of burn–up and O/M ratio typical of next–generation fast reactors (15–20 at.%, 1.94–2.00), the change in the melting temperature predicted by the code is about 100 K for 15 at.% and 40 K for a value of the O/M ratio of 1.94; see Fig. 2.

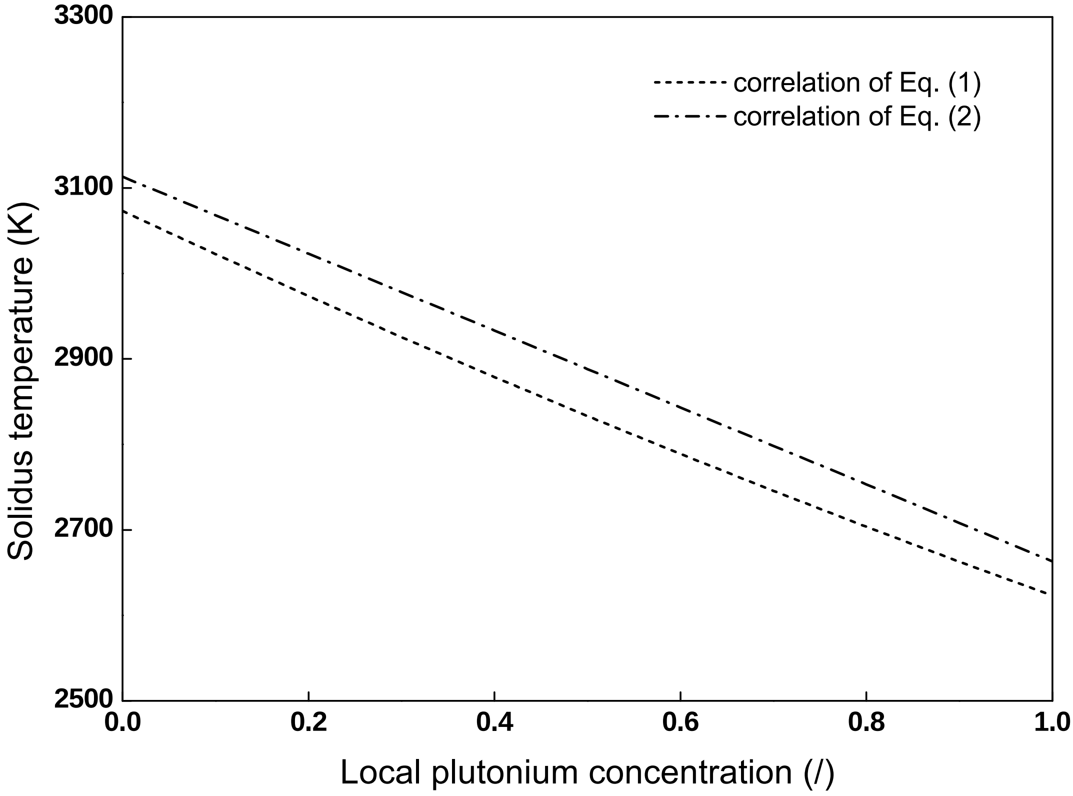

Figure 1: Predictions of solidus temperature for a stoichiometric MOX

3 EXPERIMENTAL FINDINGS ON SOLIDUS TEMPERATURE

The initial method used for the determination of the effect of burn–up was the V–shaped filament technique, afterwards, most of the data was obtained by means of the thermal arrest method [3]. In this technique, specimens are enclosed in a tungsten capsule sealed in high vacuum to overcome the sources of inaccuracies envisaged in the previous methods: the high oxygen pressure and the changes in stoichiometry occurring during the measurement [3]. The results of these experiments proved to be in agreement with the V–shaped results confirming a negligible change in the solidus temperature up to 43 GWd/t; the concentration of plutonium oxide was 18 wt.% [3].

The effect of burn–up based on the use of the V–shaped filament method had suggested a negligible effect up to 80 GWd/t for a 20 wt.% presence of plutonium [3]. At the same values of plutonium concentration (18–20 wt.%), in a burn–up domain extended up to 110 GWd/t, it was noted a slight decrease of the melting temperature beyond 50 GWd/t [4]. Based on the ideal solution theory and the results of Adamson, a model for the calculation of melting point was proposed; see § 4.1 [4,10]. The model predicts a decrease of the solidus temperature under irradiation with a rate of 7.3 K per at.% [4].

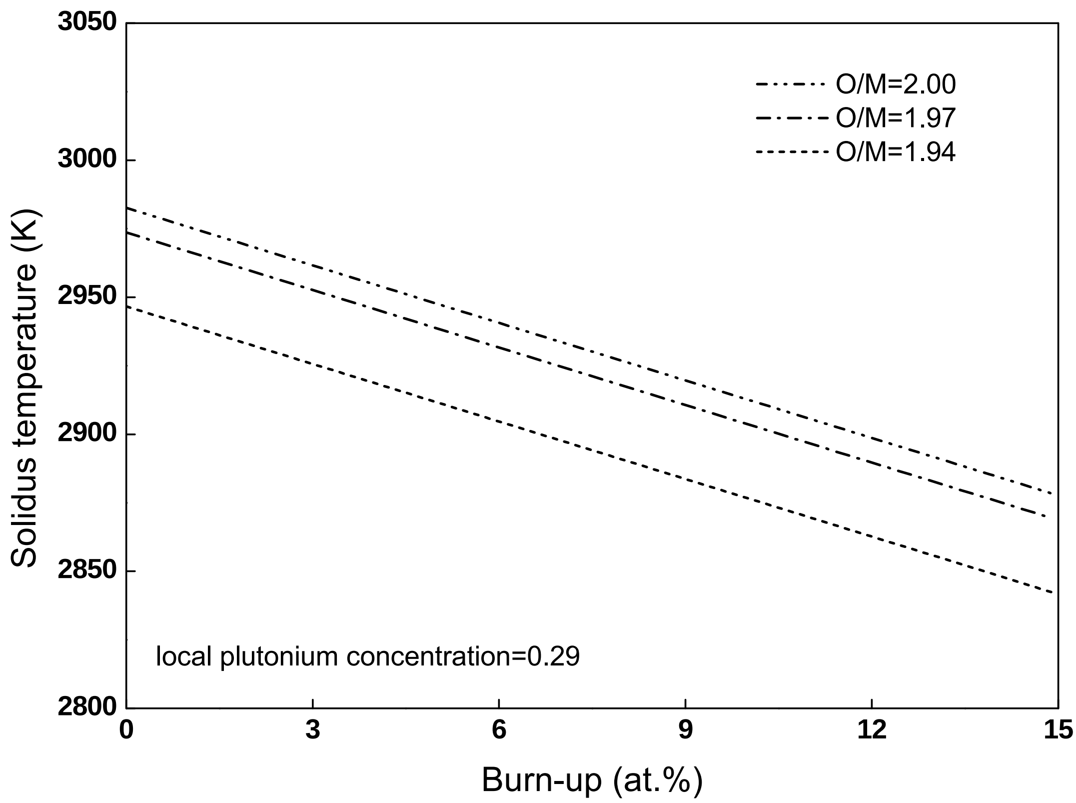

Figure 2: Predictions of burn–up and O/M ratio effects: correlation of Eq. (2)

Measurements performed for a PuO_2 concentration in the range 25–29 wt.%, O/M 1.93–1.98 and burn–up up to 124 GWd/t were presented in [5]. In particular, for a concentration of 29 wt.%, the solidus temperature was estimated to be linearly decreasing at a pace of 5 K per 10 GWd/t [5]. In the same reference [5], the negligible dependence on burn-up reported in [3,4] was questioned suggesting to reconsider the analysis at low plutonium concentrations. The driving phenomenon of this behaviour was argued to be not only the accumulation of fission products in the matrix, in fact, theoretical investigations proved to underestimate the results of experiments with an evaluation of this contribution in the term of 2 5 K per 10 GWd/t [5]. In [5], some interactions between the capsule and the specimens were reported as well.

Measurements on specimens irradiated up to 124 GWd/t in the JOYO reactor were presented in [6]. The authors developed a model through a regression analysis of the experimental data considering the actual composition of the samples at measurement through numerical simulations [6]. The correlation predicts an effect of burn−up on the depression of solidus temperature of about 6 K per 10 GWd/t with a rate decreasing at high burn−up [6]. This prediction was close to the evaluation in [4] and higher than the analytical evaluations presented in [5].

To explain the discrepancies, it was suggested that the non−ideal behaviour was caused by the formation of eutectic phases with less soluble fission products [6]. In agreement with an ideal solution behaviour, the model accounts for the presence of plutonium with a coefficient similar to that found by Adamson [6,10]. For minor actinides, the model predicts a decrease of about 10 K per wt.%. The effect of the O/M ratio was considered negligible except for un−irradiated or low burn−up fuel (< 50 GWd/t) where a nearly constant behaviour of the solidus temperature was found [3,4]. The model was refined extending its domain of application through measurements on SIMFUEL up to 250 GWd/t where the saturation of burn−up effect was confirmed [7]. The correlations were further improved with new experimental results and refined experimental data, in particular by introducing the effect of the O/M ratio [8]. The correlations of this model are presented in § 4.3.

Lyon and Baily had investigated the effect of the concentration of PuO_2 on the values of solidus temperature in stoichiometric MOX fuel by means of the thermal arrest method with a tungsten capsule sealed in argon atmosphere [9]. The results they presented were markedly consistent with the hypothesis of an ideal solution; the importance of the calibration in the high temperature region was also highlighted [9].

The correlation of the melting point recommended by the code is based on these experimental results [2]. The model proposed in [10] is also based on a re-evaluation of the measurements discussed in [9]. The work carefully investigated the effect on the solidus temperature due to the accumulation of soluble fission products under irradiation [10]. The numerical evaluations confirmed a depression of the solidus temperature of the order of 24.5 K for 10 at.% a value based on the hypothesis of ideal solution and plutonium concentration of 20 wt.% [10]. Insoluble fission products, usually in small quantities, were judged to be of minor importance in the melting of the fuel bulk assumed to be driven by the solidus temperature [10].

The dependence of the solidus temperature on the O/M ratio of MA−bearing MOX was investigated in [11]. The studied concentration of minor actinides was 4 wt.% and 2 wt.% the concentration of fission products [11]. The study reported that, for a concentration of plutonium of 30 wt.%, the melting temperature tends to decrease with an increasing O/M ratio [11].

While in ref. [12] a good agreement with the data published in [9] was noted, the experimental results for high plutonium concentration (30 and 40 wt.%) proved to be affected by a reaction of the specimens with the W−capsule [12]. As to the effect of stoichiometry, the temperature of the thermal arrest decreased with the increase of the O/M ratio [12]. The following study proved the reaction of the specimens with the tungsten capsule and proposed to overcome this limitation by using a rhenium capsule [13]. For a concentration of plutonium of 40 wt.%, the experimental result obtained by using the Re−capsule showed a value 100 K higher than previously measured [13]. The fitting of the experimental data according to an ideal solution model suggested a value of the PuO_2 melting point of 2843 K, i.e. 200 K higher than previous results, and an impact of the americium content of the order of 4 K per wt.% [13].

As to the effect of burn–up, a comparison of Re− and W−capsule measurements was presented in [14]. The range of the experimental data spans up to 112.5 GWd/t for a plutonium concentration of about 30 wt.% [14]. In this domain, the authors reported a linear decrease of the melting point at a rate in good agreement with published models; measurements performed using a Re−capsule showed absolute values higher by about 30 K [14]. The solubility limit of fission products confirmed to be a possible bound in the decrease of melting point at very high burn–up [14]. A MOX fuel initially containing 3 wt.% of americium and irradiated at 150 GWd/t was simulated and its melting temperature measured adopting the thermal arrest technique with a Re−capsule [15]. The obtained result was within the uncertainties of previous published results [15]. The authors of [16], dealing with low contaminated un−irradiated MOX fuel, suggested an impact of minor actinides on the solidus temperature of the order of 2−4 K per wt.%. The O/M ratio proved to diminish the value of the solidus temperature by about 30 K moving from 1.94 to stoichiometry [16].

To overcome the problems due to the interaction of the specimens with the capsule, the application of the laser heating technique was proposed. The measurement of the melting point of PuO_2 by means of the laser heating method gave a value of 3017 K, much higher than previously assumed [17]. On the track of this result, the melting temperature of MOX fuel with high plutonium concentrations was carried out [18]. The results, consistent with the findings of [17], showed, in general, values higher than presented in [9]. This data highlighted the need to redefine the phase diagram of the binary system UO_2–PuO_2 in the high plutonium concentration domain [18].

4 MODELS AVAILABLE IN LITERATURE

This section briefly presents the models discussed in the report. The name given to each model is referring to the first in the list of authors of reported references.

4.1 Komatsu model

The model by Komatsu calculates the value of solidus temperature according to the following formula with a dependence on plutonium, burn−up and stoichiometry [4]. The solidus temperature of UO_2 is assumed to be 2865 °C. The *X* parameter is the concentration of plutonium where burn−up (*BU*) is expressed in at.%.

$$T_{m,sol} = \frac{1-0.2388(2-O/M)}{1+0.1811Y-0.011Y^2} T_m(UO_2) \quad (5)$$

$$Y = X + 0.016BU \quad (6)$$

4.2 Adamson model

The model by Adamson deals with un–irradiated and stoichiometric MOX through a polynomial expression dependent on the concentration of plutonium [9,10]. In presented results, the predictions of the model were refined through a multiple regression of the numerical evaluations of the impact of burn−up [10]. The correlation by Adamson (K) is:

$$T_{m,sol} = 3120.0 - 655.3c_{Pu} + 336.4{c_{Pu}}^2 - 99.9{c_{Pu}}^3 \quad (7)$$

4.3 Konno model

The model by Konno expresses the dependence of the solidus temperature on plutonium concentration, O/M ratio, stoichiometry and americium concentration [6–8].

$$T_{m,sol} = 3138 - 497c_{Pu} + 60{c_{Pu}}^2 - \Delta T_{O/M} - \Delta T_{Am} - \Delta T_{bu} \quad (8)$$

where:

$$\Delta T_{bO/M} = (1000 - 2850c_{Pu})(2 - O/M) \quad (9)$$

$$\Delta T_{Am} = (1206 - 782c_{Pu})c_{Am} \quad (10)$$

$$\Delta T_{bu} = (1.06 - 1.43c_{Pu})bu - 0.0008((1.06 - 1.43c_{Pu})/0.66)^{1.5} bu^2 \quad (11)$$

The model is valid in the following domains:

- Plutonium concentration 10–40 wt.%.
- Americium concentration <2 wt.%.
- O/M ratio 1.94–2.00.
- Burn−up 0–250 GWd/t.

5 REFERENCE EXPERIMENTAL DATASET

Table 1 resumes some information regarding the experimental dataset applied in the assessment of the investigated models. Measurements on SIMFUEL presented in [7,15] were not used in this study.

6 RESULTS AND DISCUSSION

The predictions of the model recommended by the code are compared with the related experimental data in Fig. 3. The predictions show deviations mostly of the order of ±3%. Based on the assumption that the effect of burn–up is in good agreement with the literature review, noting that the impact of MAs and O/M ratio turned out to be limited in the predictions of selected experimental points, a deeper consideration for the effect of the concentration of plutonium is needed. As shown in Fig. 3, the accuracy of the correlation is close to the experimental uncertainties considering the points with plutonium concentrations lower than 20 wt.%. In agreement with the considerations leading to the use of a Re–capsule, eq. (2) in general underestimates the measurements by Kato while the data by Hirosawa confirms a nearly constant underestimation in the considered burn–up domain. Much higher deviations were found for the data published in [18].

Preliminary conclusions drawn from this assessment could suggest that the recommended code correlation shows a fairly good description of the effect of burn-up and plutonium concentration up to 20–25 wt.% thereafter the underestimation is more significant and, in general, increasing with the concentration. The description of the effect of the O/M ratio is not in agreement with recent results where the solidus temperature is decreasing with the increasing of the O/M ratio.

Table 1: Description of significant information of the adopted experimental dataset

Reference	No. of points	BU GWd/t	Pu %	MAs %	O/M –	Technique*
Tachibana [3,8]	4	8.2–43	18	0.3	1.99–2.00	TA–W
Komatsu [4,8]	4	51.4–110.9	16–17	0.9	1.99	TA–W
Konno [6]	21	0.0–124.0	27–29	0.4–1.2	1.95–1.98	TA–W
Konno [8]	6	33.6–47.1	17	0.5	2.00	TA–W
Lyon [9]	10	0.0	0–100	0.0	2.00	TA–W
Kato [13]	22	0.0	12–60	0.3–3.3	1.94–2.00	TA–W/–Re
Hirosawa [14]	12	0.0–112.5	28–30	0.0	1.96–1.98	TA–Re
Kato [16]	4	0.0	30	3.2	1.96–1.98	TA–Re
Manara [18]	4	0.0	75–100	0.0	1.98	LH

*(TA=thermal arrest; LH=laser heating)

Table 2 and Fig. 4 show standard deviation and bias of the calculated predictions. According to the results from eq. (2), see Fig. 3, the values published in [18] were not taken into account in the comparison. The analysis is therefore referring to a plutonium domain with concentrations lower than 60 wt.%.

Table 2: Values of parameters selected for the comparison of models

Model	No. of points	Std. Deviation	Bias
Correlation of Eq. (1)	83	90.9	-84.3
Correlation of Eq. (2)	83	46.7	-33.6
Komatsu	83	42.7	-22.0
Adamson	83	50.6	-34.0
Konno	70	30.8	-16.6

The comparison confirms that eq. (1) is markedly conservative with the highest deviation where the model by Konno has the best performance with deviations of the order of the experimental uncertainties; see Fig. 4. All models have a negative bias with the Konno model presenting the less pronounced underestimation. Eq. (2) and the Adamson model are in fair agreement as they were derived from the same experimental data. Though being different, Konno and Komatsu are both well–performing. Noting that the dependence on the plutonium concentration in eq. (2) is similar to that of Konno, the better performance of the latter could be explained by the high fraction of the experimental dataset used for its development.

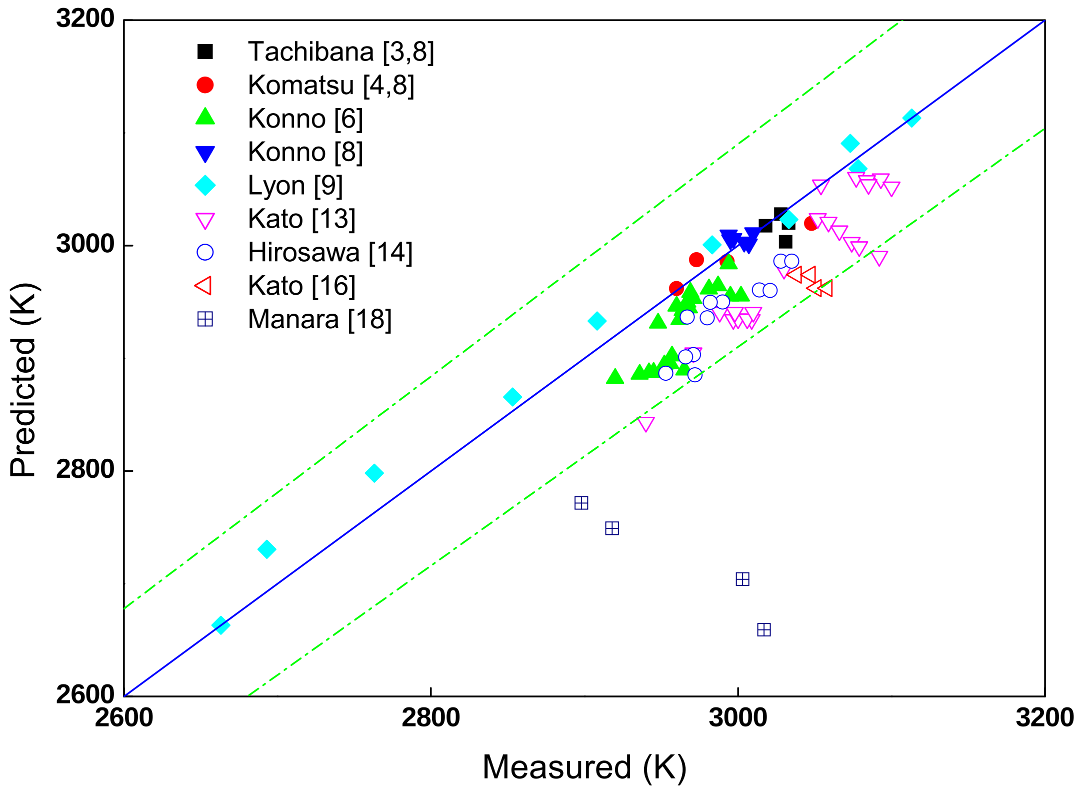

Figure 3: Predictions of eq. (2) vs. experimental dataset (dashed lines ±3% accuracy)

The comparison confirms that the Konno model could be of particular interest for the code because it considers the presence of minor actinides (americium) and the saturation of the burn–up effect. Nevertheless, the Komatsu model achieved results close to the Konno model without limitation in the domain of its parameters. It could therefore be of interest in case the redistribution under irradiation leads to local plutonium concentrations above 40 wt.%.

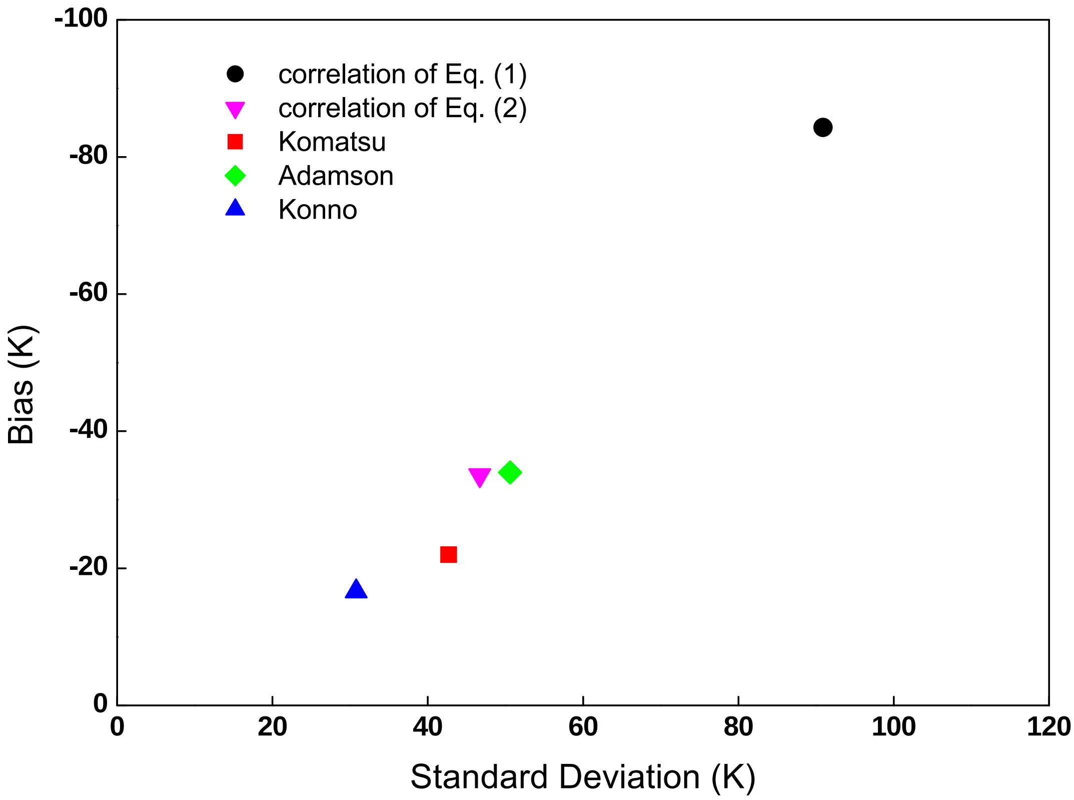

Figure 4: Comparison of models (Standard deviation, Bias)

7 CONCLUSIONS

The document presented the main experimental findings and models for the evaluation of the solidus temperature of MOX fuel for FBRs. The recommended model of TRANSURANUS proved to be fairly accurate for low plutonium concentrations while, according to recent published results, an underestimation of its predictions was noted at higher concentration of plutonium. The burn-up effect is consistent with literature results while the effect of the O/M ratio is not in agreement with recent modelling. A preliminary analysis of this comparison confirmed the accuracy of the Konno and Komatsu models. The latter is of interest for significant values of actinides redistribution.

These preliminary results could be of interest for future sensitivity studies to discuss the impact of the correlations on the margin to melting, a key parameter in the safety analysis.

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