

## Thermal Cycling Growth of Metallic Uranium Alloys

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### Abstract

For the development of high power target elements for the German spallation neutron source (SNQ) thermal cycling growth of  $\alpha$ - and  $\gamma$ -uranium alloys were studied up to 400,000 cycles as a function of the lower cycling temperature  $T_0$  and the cycling amplitude  $\Delta T$ . The growth rates obtained for the  $\alpha$ -uranium alloys were not constant for given  $T_0$ ,  $\Delta T$  and alloy composition, but could be subdivided in an initial growth rate  $G_0$  at cycle numbers smaller than 50,000, a lower permanent growth rate  $G_p$  above 100,000 cycles and a transient growth rate  $G_T$  in between.

The initial growth rate  $G_0$  increases strongly with increasing  $T_0$  and  $\Delta T$ . Compared to  $G_0$ , the dependence of the permanent growth rate  $G_p$  on  $T_0$  and  $\Delta T$  is less.

In contrast to  $\alpha$ -uranium, no thermal cycling growth could be found of the metastable  $\gamma$ -phase of an UMo10 W/o alloy.

### 1. Introduction

Neutron scattering is increasingly used to study material properties of condensed matter. As the number of fission reactors available as neutron sources for research purposes as well as their neutron flux densities are limited, new options are studied to produce high neutron fluxes. The spallation neutron source (SNQ) proposed for construction at KFA Juelich, will use a time averaged 5 mA current of 1.1 GeV protons to produce neutrons by spallation of uranium as target material, following a start-up period with a tungsten target. Although there is much less heat per neutron dissipated in a spallation target than in a fission reactor core, the total heat deposited in an uranium target of a 5 mA, 1.1 GeV facility is still about 10 MW. With a beam diameter around 4 cm and a penetration depth of 40 cm, limiting the target temperature becomes a major problem. To reduce the heat load as well as the radiation damage for a given target area by a factor of 200, a rotating target with a diameter of 2.5 m is used. The target wheel contains about 8500 clad cylindrical uranium pins (20 x 100 mm) separated by 1 mm gaps through which the cooling water is flowing. An angular velocity of 0.5 Hz is chosen, so that consecutive proton pulses (separated by 10 ms) will hit adjacent target areas, allowing 2 s (or one rotation of the target wheel) for the heated target area to cool down.

Although the target wheel concept reduces the lower operation temperature  $T_0$  of an uranium target to less than 200 °C and the temperature increase  $\Delta T$  per proton pulse under 100 K, the endurance resistance of the target material during the anticipated operation time of two years (about  $2 \times 10^7$  cycles) has to be high. Therefore programs were started to study the performance of uranium alloys under thermal cycling and, subsequently, under mechanical load. Results of the thermal cycling program, varying lower cycling temperature  $T_0$  and  $\Delta T$  are presented in this paper.

## 2. Experimental

To study the effect of thermal cycling upon uranium alloys, two methods were used:

- a. Sets of up to 12 samples for each  $\Delta T$  were mechanically transferred between thermostates with silicon oil at different temperatures. The holding time (immersion time) at the lower and upper temperatures was 15 s each, the transfer time of the samples  $2 \times 2$  s. Up to 6 thermostates were placed in a glove box, flooded with nitrogen.
- b. A single sample was periodically heated in a 4 kHz medium frequency induction furnace and cooled by a silicon oil containing cooling circuit.

Two allotropes of uranium alloys were under investigation:

$\alpha$ -uranium: this orthorhombic phase shows a strong anisotropic thermal expansion coefficient and thus a pronounced irreversible swelling under thermal cycling.

$\gamma$ -uranium: in contrast to the  $\alpha$ -phase no cycle dependent swelling is observed in the metastable body-centered cubic  $\gamma$ -phase. But it has to be shown, that no phase transition occurs under operation, i. e. thermal cycling, alternating mechanical load and radiation.

Samples: The type of uranium alloys investigated are listed in Table 1, the compositions given in weightpercent. With the exception of the  $\gamma$ -phase of UM010, all other uranium alloys are in the modification of the  $\alpha$ -phase. "Adjusted uranium" used as stationary target at the APNS of Argonne National Laboratory, USA, and the SNS of Rutherford Appleton Laboratory, UK, contains 470 ppm C, 250 ppm Fe and 250 ppm Si.

The samples cycled were cylinders, 9 mm in diameter and 50 mm in length for dimensional measurements as well as sets of discs (9 x 2 mm) for metallographic investigations.

Evaluation: After given cycle intervals, length and weight of the samples were measured. The changes in length were corrected for sample corrosion, assuming that the weight loss of the samples after cycling is due to an uniform corrosion at the sample surface.

The thermal cycling growth rate (per cycle) was determined in three different ways:

- a. by calculating the "integral growthrate"  $G_I = \mathcal{E} / n$  from the total relative change in sample length  $\mathcal{E} = (l_n - l_0) / l_0$  at a given cycle number  $n$ . ( $l_0$ : initial sample length).
- b. by calculating the mean of the growth rates for given cycle number intervals and
- c. by calculating the growth rate from the slope of the fitted  $\mathcal{E}$ -curve.

From the data determined according methods b and c for cycle numbers above 100,000, the mean was calculated and referred to as permanent growth rate  $G_p$ .

## 3. Results

In general, the thermal cycling growth rates of the  $\alpha$ -uranium alloy samples were not constant at given  $T_0$ ,  $\Delta T$  and alloy composition but showed three different categories depending on the number of thermal cycles accumulated.

Region I (initial growth rate  $G_0$ ; 0 to about 50,000 cycles) shows strong dimensional changes. Except for the lowest  $\Delta T$  (10 K) and  $T_0 = 55^\circ\text{C}$ ,  $G_0$  is high.

Region II (transient growth rate  $G_T$ , 50 to 100,000 cycles). In this region samples tend to shrink, which leads to small or negative values for  $G_T$ .

Region III (permanent growth rate  $G_p$ , above 100,000 cycles). In general the growth rate  $G_p$  measured at cycle numbers above 100,000 up to the maximal cycle number of 420,000 accumulated to date, is much smaller than the initial growth rate  $G_0$ . As the main interest of this study is centered on the material behaviour at high cycle numbers, the change of the growth rate  $G_0$  in region I was not studied in detail. But the contribution of the dimensional changes at low cycle numbers to the integral changes of a sample at the maximal cycle number achieved in this study (i. e. 420,000) can be substantial. As the integral growth rate  $G_I$  is calculated from the total dimensional changes at a given cycle number  $n$ , whereas the permanent growth rate  $G_p$  represents the dimensional changes in region III only, distinct differences between  $G_I$  and  $G_p$  were found. In Table 1,  $G_I$  and  $G_p$  for different uranium alloys are listed as a function of the lower cycling temperature  $T_0$  and the cycling amplitude  $\Delta T$ .

Variation of the growth rates at  $T_0 = 55^\circ\text{C}$ : Two examples for the typical results at  $T_0 = 55^\circ\text{C}$  are shown in Figs. 1 and 2. The variation of the integral growth rate  $G_I$  of denaturated  $\alpha$ -uranium for  $\Delta T = 10, 35$  and  $65$  K as a function of the number of thermal cycles is illustrated in Fig. 1 at the left side, at the right side, the corresponding curves for  $\mathcal{E} = \Delta l/l_0$  are shown. The analogous diagrams are displayed in Fig. 2 for an UMo5 alloy. All uranium alloys with  $\alpha$ -phase showed in region I negative values for  $G_0$  at  $\Delta T = 10$  K and high values for  $G_0$  at  $\Delta T = 35$  and  $65$  K. As it is likely, that the maximum of  $G_0$  lies at cycling numbers smaller than that of the first measurement (i. e. 40,000 cycles), values for  $G_0$  are not listed in Table 1.

In region II (50 000 to about 100 000 cycles), the integral growth rate  $G_I$  decreases for  $\Delta T = 35$  and  $65$  K, and the transient growth rate  $G_T$  in cycle number interval region II may become even negative.

For region III, for cycles above 100,000 the integral growth rate  $G_I$  remains nearly constant for a given  $\Delta T$ , but yields higher values for higher  $\Delta T$ . But as the slopes of the  $\mathcal{E}$ -curves at the right side of Figs. 1 and 2 show, this is only due to differences of  $G_0$ . The permanent growth rate  $G_p$  is nearly independent of  $\Delta T$  in the range of  $\Delta T = 10$  to  $65$  K. In contrast to this, the  $G_p$ -values at  $\Delta T = 170$  K are higher for all  $\alpha$ -uranium alloys investigated (Table 1 and Fig. 3).

Variation of the growth rates at  $T_0 = 125^\circ\text{C}$ : As an example, Fig. 4 shows the integral growth rates  $G_I$  and the relative dimensional changes at  $\Delta T = 35, 65$  and  $100$  K for uranium. The variation of  $G_I$  and  $\mathcal{E}$  with the number of thermal cycles is similar to that at  $55^\circ\text{C}$ . Only the values are higher and the maximum of  $G_0$  seems to be shifted to lower cycle numbers. By and large, the contributions of dimensional changes in region I to the total change in sample length is smaller at  $125^\circ\text{C}$  than at  $55^\circ\text{C}$ , resulting in very similar values for  $G_I$  and  $G_p$ .

In general, uranium showed the highest growth rate in comparison to  $\alpha$ -uranium alloys in all three cycle number regions, "adjusted uranium" was at the lower range. But the first results at  $T_0 = 160^\circ\text{C}$  and  $\Delta T = 35, 65$  and  $100$  K indicate, that "adjusted uranium" exhibits the highest swelling rate under these conditions.

The UMo10 samples with the metastable  $\gamma$ -phase investigated up to 420 000 cycles did not show any significant growth even at the highest  $T_0$  of 160 °C at  $\Delta T = 100$  K or the highest  $\Delta T$  of 205 K at  $T_0 = 55$  °C.

The first results of metallographic investigations of cycled samples show a relationship of the different swelling rates in the regions I to III with microstructural variations. Although UMo5 is, due to the corrosion resistance, the most interesting material of all  $\alpha$ -uranium alloys investigated, an unambiguous phase identification could not yet be achieved, as four modifications of an  $\alpha$ -phase as well as of a  $\gamma$ -phase may be present.

In region I a strong increase in optical anisotropy was observed, probably due to transformation of residues of the bcc  $\gamma$ -phase into the anisotropic  $\gamma'$ -phase, starting at grain- and phase-boundaries. In region II a densification and a rearrangement of the microstructure takes place, accompanied by a strong decrease in optical anisotropy. Region III again shows an increase in anisotropy, but to a smaller degree than in region I, corresponding to the lower values of  $G_p$  compared to  $G_0$ . At higher temperatures  $T_0$  or larger  $\Delta T$ , the specific microstructures appear at lower cycle numbers, indicating a superposition of cycling and tempering effects. In order to differentiate between these effects, samples will be isothermally annealed parallel to thermal cycling.

In earlier investigations of thermal cycling growth of  $\alpha$ -uranium by Mayfield /1/, Lloyd et al. /2/, Zegler et al. /3/ and Burke et al. /4/ the maximal number of thermal cycles was limited to about 3000. As this region is only partially covered by this study in form of  $G_0$ , a comparison of the described date for  $G_0$  with findings of other authors can be only qualitativ. But as stated in the literature, the initial growth rate  $G_0$  increases with increasing  $T_0$  as well as with  $\Delta T$ .

#### References

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- /2/ Lloyd, L. T., Mayfield, R. M., "Microstructural changes of uranium upon thermal cycling", Trans. Amer. Soc. Metals 50, 954 (1985)
- /3/ Zegler, S. T., Mayfield, R. M., Mueller, M. H., "Effects of fabrication and heat treatment variables upon the thermal cycling behaviour of uranium", Trans. Amer. Soc. Metals 50, 905 (1985)
- /4/ Burke, J. E., Turkalo, A. M., "The growth of uranium upon thermal cycling", Trans. Amer. Soc. Metals 50, 943 (1985)

Table 1: Results of thermal cycling with  $\alpha$  - and  $\gamma$  -uranium alloys

Lower Cycling Temperature To (°C)	Temperature Amplitude $\Delta T$ (K)	Uranium Alloy	Permanent Growthrate $G_p$ ( $10^{-9}$ )	Integral Growthrate $G_I$ ( $10^{-9}$ )
55	+ 10	UMo 5	$2 \pm 1$	$0 \pm 0.5$
		UZr 3.7	3 "	0 "
		UTi 0.7	2 "	0 "
		U	4 "	$2 \pm 1$
	+ 35	UMo 5	$2 \pm 1$	$2 \pm 1$
		UZr 3.7	2 "	2 "
		UTi 0.7	1 "	2 "
		U	4 "	5 "
	+ 65	UMo 5	$2 \pm 1$	$4 \pm 1$
		UZr 3.7	2 "	5 "
		UTi 0.7	2 "	4 "
		U	4 "	6 "
	+ 170	UMo 5	$13 \pm 2$	$14 \pm 2$
		UTi 0.7	12 "	12 "
		U-Adj.	11 "	10 "
		U	20 "	20 "
UMo 10		$0.5 \pm 1$	$0.5 \pm 1$	
125	+ 35	UMo 5	$10 \pm 3$	$11 \pm 2$
		UTi 0.7	11 "	11 "
		U-Adj.	11 "	9 "
		U	8 "	8 "
		UMo 10	$2 \pm 2$	2 "
	+ 65	UMo 5	$16 \pm 4$	$19 \pm 3$
		UTi 0.7	17 "	16 "
		U-Adj.	17 "	15 "
		U	17 "	20 "
		UMo 10	$-2 \pm 2$	$-4 \pm 2$
	+ 100	UMo 5	$14 \pm 4$	$13 \pm 3$
		UTi 0.7	18 "	15 "
		U-Adj.	15 "	13 "
		U	18 "	17 "
		UMo 10	$1 \pm 2$	$0.5 \pm 1$

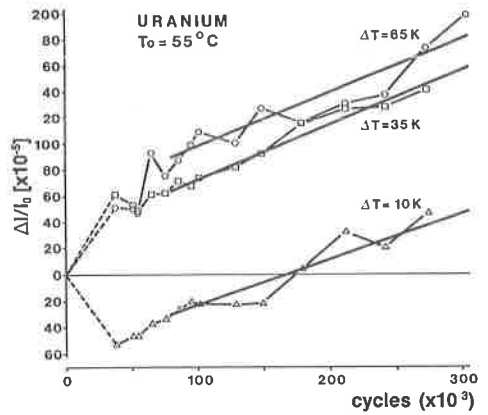
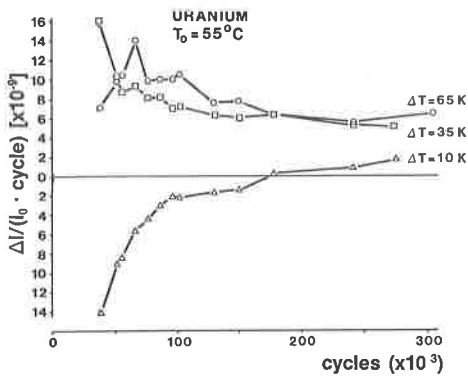


Fig. 1: Integral thermal cycling growth rate  $G_I$  (left) and relative dimensional changes  $\mathcal{E}$  (right) of denaturated uranium as a function of thermal cycles at  $T_0 = 55^\circ\text{C}$  and  $\Delta T = 10, 35$  and  $65\text{ K}$

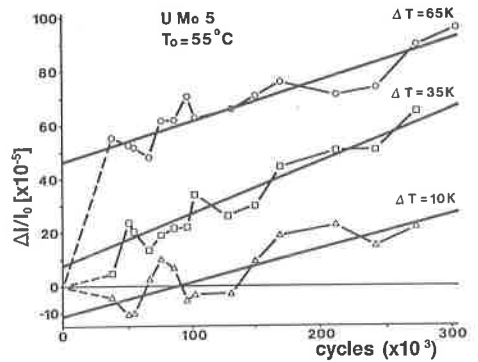
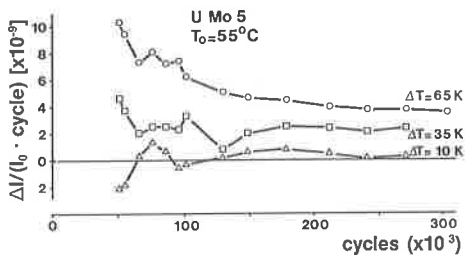


Fig. 2: Integral thermal cycling growth rate  $G_I$  (left) and relative dimensional changes  $\mathcal{E}$  (right) of an  $\text{U Mo 5}^{W/O}$  alloy as a function of thermal cycles at  $T_0 = 55^\circ\text{C}$  and  $\Delta T = 10, 35$  and  $65\text{ K}$ .

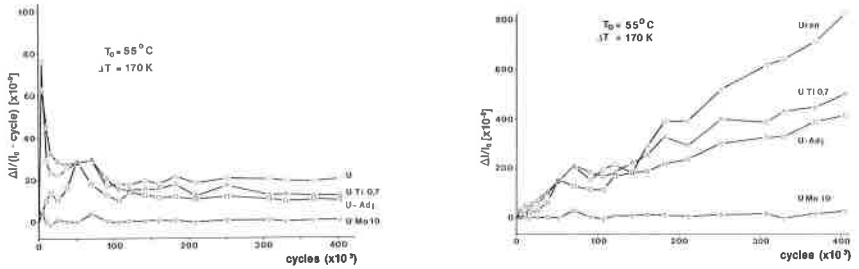


Fig. 3: Integral thermal cycling growth rate  $G_I$  (left) and relative dimensional changes  $\Delta l/l_0$  (right) of uranium, UTi.07, U-Adjusted (all as  $\alpha$ -phase) and UMo 10 ( $\gamma$ -phase) at  $T_0 = 55^\circ\text{C}$  and a  $\Delta T$  of 170 K.

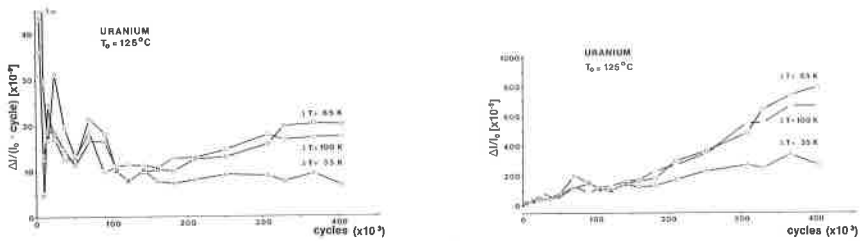


Fig. 4: Integral thermal cycling growth rate  $G_I$  (left) and relative dimensional changes  $\Delta l/l_0$  (right) of uranium as a function of thermal cycles at  $T_0 = 125^\circ\text{C}$  and  $\Delta T = 35, 65$  and 100 K.