

Thermal Cycling Growth of Uranium Alloys and Fabrication
of SNQ Target Pins

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Summary

Targets of neutron spallation sources are exposed to cyclic thermomechanical load induced by the pulsed proton beam. For the development of high power target elements, the cycling growth rates of α -uranium and α -uranium alloys were studied up to 550,000 cycles as a function of the lower cycling temperature T_0 and the cycling amplitude ΔT . The growth rates obtained for α -uranium were not constant for given T_0 , ΔT and material composition. Depending on the number of thermal cycles accumulated, the growth rate G can be classified in four categories: At cycle numbers $n < 50,000$, the G values are high and strongly dependent on T_0 and ΔT . In the cycle number intervals 50,000 to 100,000 and about 450,000 to 550,000 most samples tend to shrink, which leads to negative growth rates. At cycle numbers $100,000 < n < 450,000$ the growth rate is hardly dependent on ΔT and smaller than the initial growth rate. For UM10 alloys with the bcc γ -phase, no significant dimensional changes during thermal cycling were observed.

For the production of clad uranium target elements, several fabrication techniques were tested to ensure high thermal conductivity at the uranium/cladding interface during target operation. Based on the material properties of the as-produced target elements, hot isostatic pressing of UM10 spallation material and Zry-2 cladding gave the best result of all target element variations investigated.

Introduction

The UM010 alloy is expected to have the best performance of the uranium based spallation materials under thermomechanical cyclic load for the following reasons: a) lack of thermal cycling growth of the bcc γ -phase in contrast to the orthombic α -phase and b) lower corrosion rates in water by a factor of 100 to 2000 compared to uranium.

As the γ -phase is metastable at temperatures below 774 °C, a phase transformation may occur during target operation at temperatures up to 320 °C. Therefore the γ -phase stability under thermal cycling as well as the effect of lower cycling temperature T_0 and cycling amplitude ΔT (Table 1) on the thermal cycling growth of α -uranium and α -uranium alloys were investigated.

Lower Cycling Temperature T_0 (°C)	Cycling Temperature Amplitude ΔT (K)
55	+ 10, + 35, + 65, + 100, + 170, + 205
125	+ 35, + 65, + 100
160	+ 35, + 65, + 100

Table 1: Lower cycling temperature T_0 and temperature amplitude ΔT of the α -uranium cycling program.

Experimental

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

- a) Sets of up to 12 samples for each ΔT 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 2×2 s. Up to 6 thermostates were placed in a glove box, flushed 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.

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. The materials investigated were uranium, UMo5, UZr3.7, UTi0.7 and "adjusted uranium" with 470 ppm C, 250 ppm Fe and 250 ppm Si (all as α -phase) and UMo10 (as γ -phase).

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" $G_I = \xi / n$ from the total relative change in samples length $\xi = (l_n - l_0) / l_0$ 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 ξ -curve.

For optimization of integral target elements, different cladding procedures were applied: hot isostatic pressing (HIP), extrusion, shrink fitting and soldering. UMol0 was used as spallation material with and without a nickel bonding. The Ni coating was deposited either by electrolytic coating or PVD. As cladding material Zry-2, AlMg3 or AlMgSi0,5 was chosen.

The quality of the uranium/(bonding)/cladding interface, which is important for high thermal conductivity, was investigated by optical inspection of metallographic sections and electron microprobe analysis (MPMA) as well as measurements of shearing and tensile strength.

Results

a) Thermal cycling

Uranium and uranium alloys with α -phase: Depending on the number of cycles accumulated, different swelling mechanism occur during thermal cycling at given T_0 , ΔT and material composition, leading to varying growth rates. In the cycle number intervall investigated (0 to 550,000 cycles), four categories of growth rates can be distinguished:

- G_0 ($n < 50,000$) is very sensitive to cycling parameters and increases with increasing ΔT (Fig. 1) and T_0 (Figs. 1 and 3), which is in agreement with literature data
- G_1 ($50,000 < n < 100,000$) is mostly negative
- G_2 ($100,000 < n < 400,000$) is smaller than G_0 and hardly dependent on ΔT , as can be concluded from the similarity of the slopes of the fitted ξ -curves in Fig. 2 and the same values for the growth rates G_1 in Fig. 3.
- G_3 ($400,000 < n < 550,000$) is negative for most samples as indicated by the negative slope of the ξ -curve above 400,000 cycles in Fig. 4.

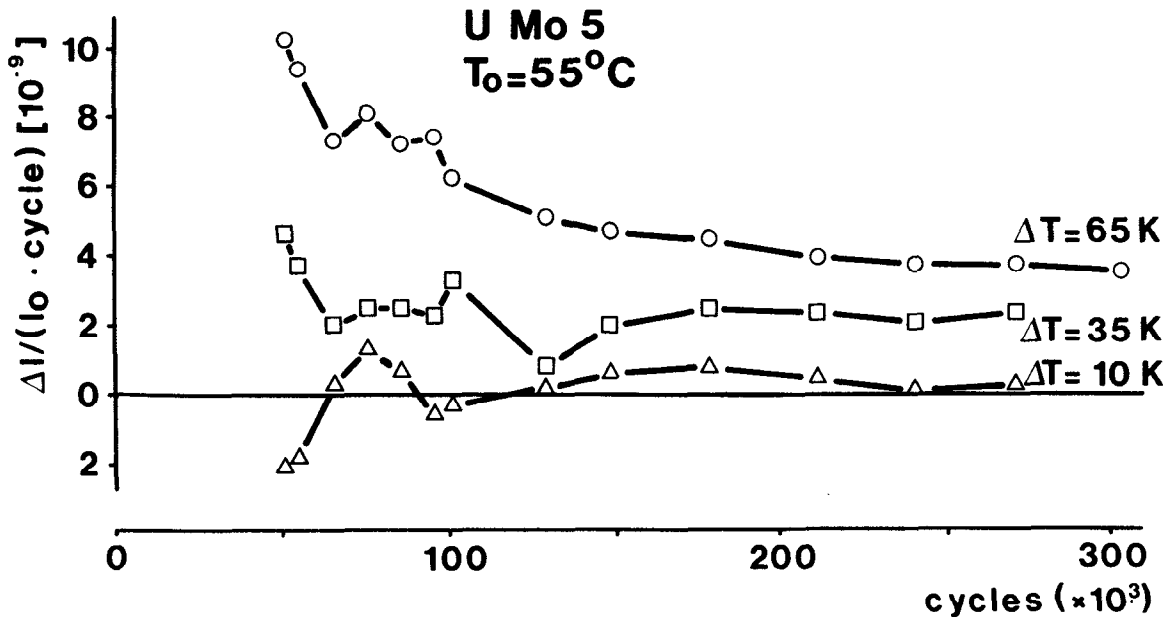


Fig. 1: Cumulative thermal cycling growth rate (per cycle) vs. cycle number of UMo5 at a lower temperature T_0 of 55 °C and $\Delta T = 10, 35$ and 65 K

In general α -uranium shows a higher growth rate than the α -uranium alloys investigated. From these, "adjusted uranium" yields the lowest G-values at low T_0 and small ΔT , but the highest growth rates at high T_0 and ΔT .

For the application of α -uranium alloys as target material it is important, that although G_0 is high and proportional to T_0 and ΔT , the thermal cycling growth rate at high cycle numbers is much smaller or even negative and nearly independent of ΔT . The total dimensional changes of the samples measured at a cycle number of 550,000 was, except for uranium, at least a factor of 10 to 30 smaller than these calculated from the G_0 values, assuming a constant initial growth rate up to the above mentioned cycle number.

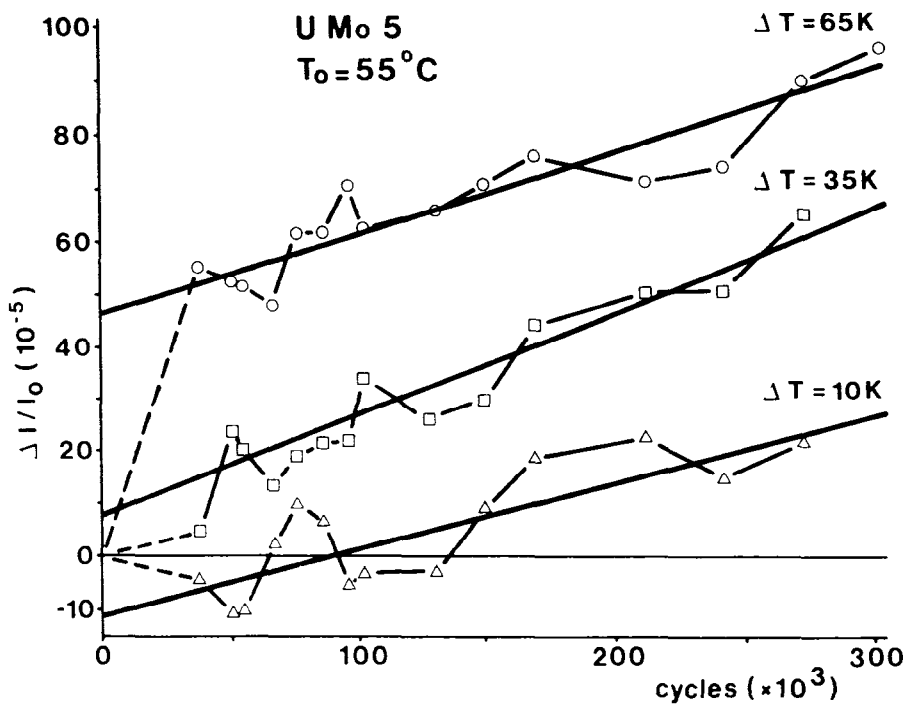


Fig. 2: Relative dimensional changes $\epsilon = \Delta l/l_0$ vs. cycle number of UMo5 at $T_0 = 550^\circ\text{C}$ and $\Delta T = 10, 35$ and 65K

The UMo10-samples with the metastable bcc γ -phase did not show any significant dimensional change even at the highest T_0 of 160°C ($\Delta T = 100\text{K}$) or the largest ΔT of 205K ($T_0 = 55^\circ\text{C}$)

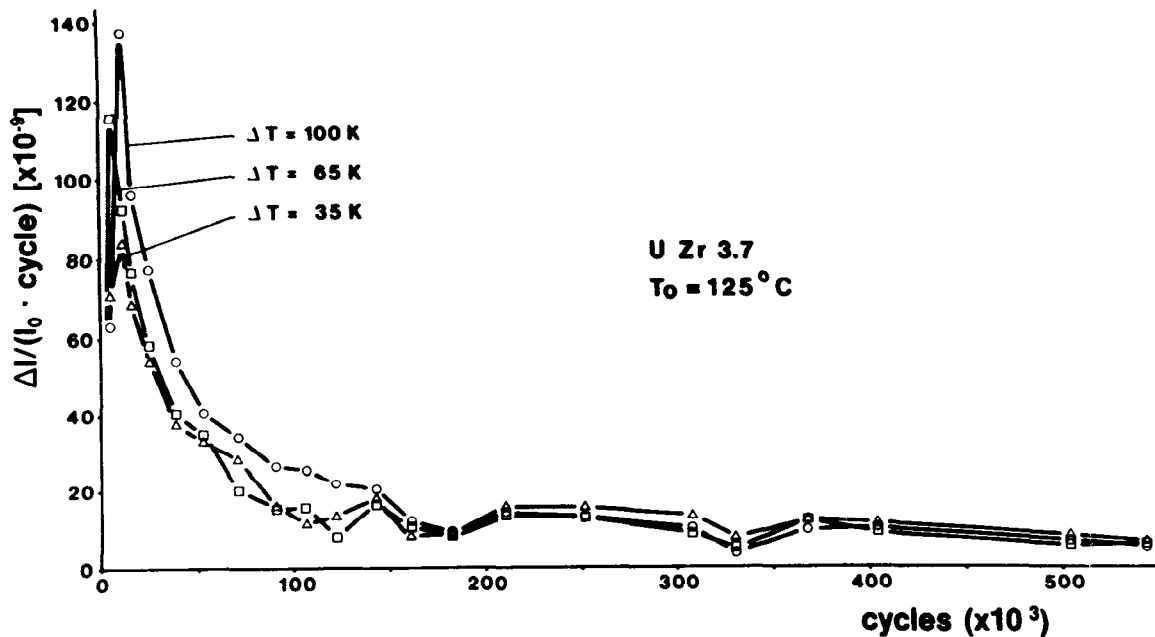


Fig. 3: Cumulative thermal cycling growth rate vs. cycle number of UZr3.7 at $T_0 = 125^\circ\text{C}$ and $\Delta T = 35, 65$ and 100K

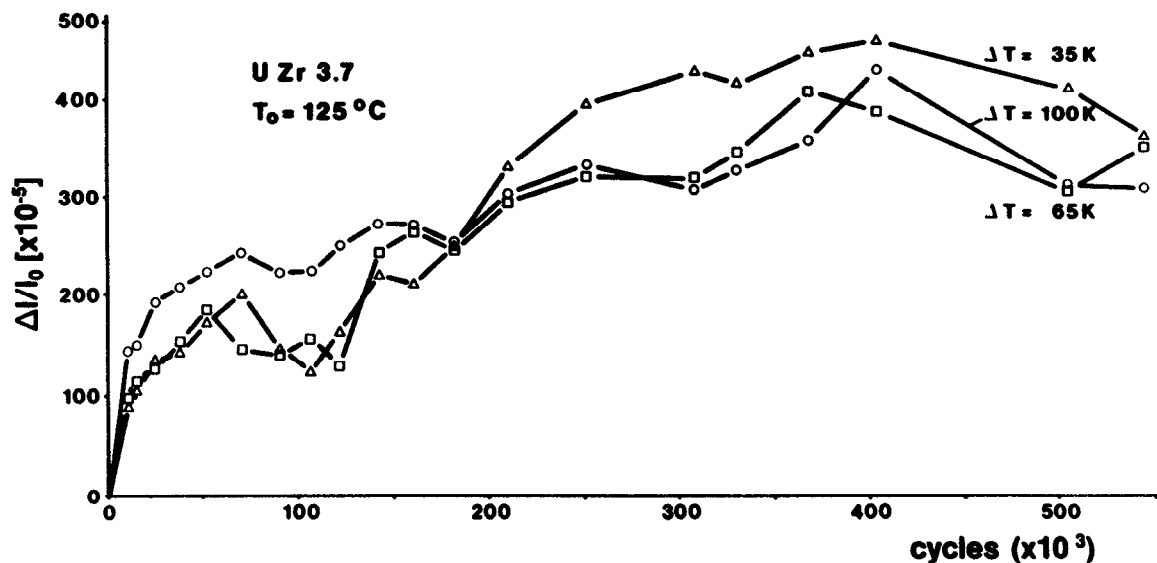


Fig. 4: Relative dimensional changes $\xi = \Delta l/l_0$ vs. cycle number of UZr3.7 at $T_0 = 125 \text{ }^\circ\text{C}$ and $\Delta T = 35, 65$ and 100 K

With a time at temperature of 15 s per cycle each at the upper and lower temperature, the equivalent temperature treatment during thermal cycling was for example for the samples at $T_0 = 160 \text{ }^\circ\text{C}$ and $\Delta T = 100 \text{ K}$ about 2,500 h (100 d) at $160 \text{ }^\circ\text{C}$ and at $260 \text{ }^\circ\text{C}$ respectively.

Samples annealed at $550 \text{ }^\circ\text{C}$ for 1 to 330 h prior to thermal cycling exhibit an increase in thermal cycling growth with initial annealing time, in qualitative agreement with the degree of γ to $\alpha + \delta$ phase transition as seen in the corresponding metallographic sections after isothermal annealing.

b) Target element fabrication

For the optimization of the UMol0/cladding interface, the thickness of the nickel bonding (0 to 40 μm) and the cladding material (Zry-2, AlMg3, AlMgSi0.5) as well as the technique for the bonding deposition and the cladding procedure was varied. Hot isostatic pressing (HIP) showed the best results in encapsulating the UMol0 spallation material, independent whether a Ni-bonding was used or not. In contrast to the other variants

(UMo10/Ni/Zry-2, UMo10/AlMg3, UMo10/AlMgSi0.5, UMo10/Ni/AlMg3, UMo10/Ni/AlMgSi0.5), hot isostatic pressing of UMo10/Zry-2 (without a Ni-bonding) did not lead to numerous intermetallic phases at the UMo10/cladding interface as in the case of the other variants.

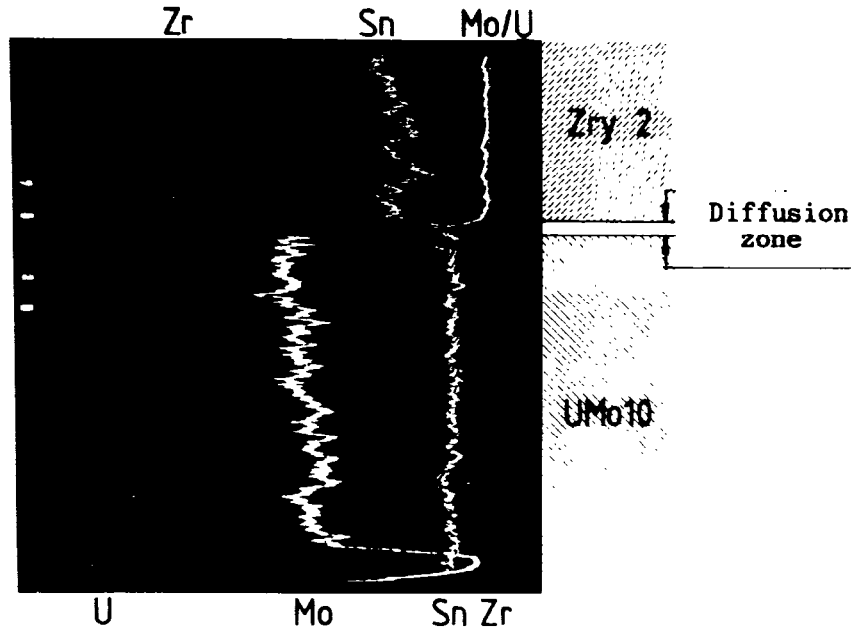


Fig. 5: MPMA linescans of the HIP-interface UMo10/Zry-2. HIP temperature: 800 °C (6 h) (900x).

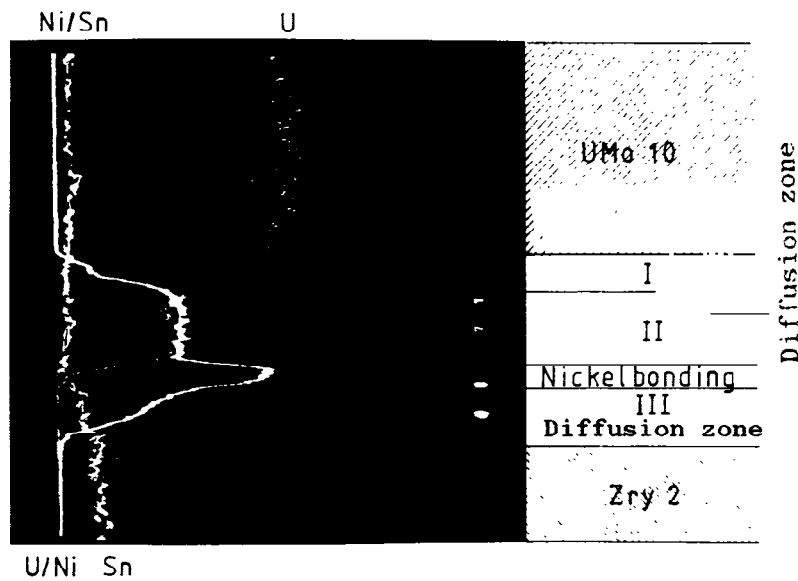


Fig. 6: MPMA linescans of the HIP-interface UMo10/Ni/Zry-2. HIP temperature: 700 °C (6 h). Deposition of Ni-bonding by electrolytic coating; initial layer thickness: 20,um (900x)

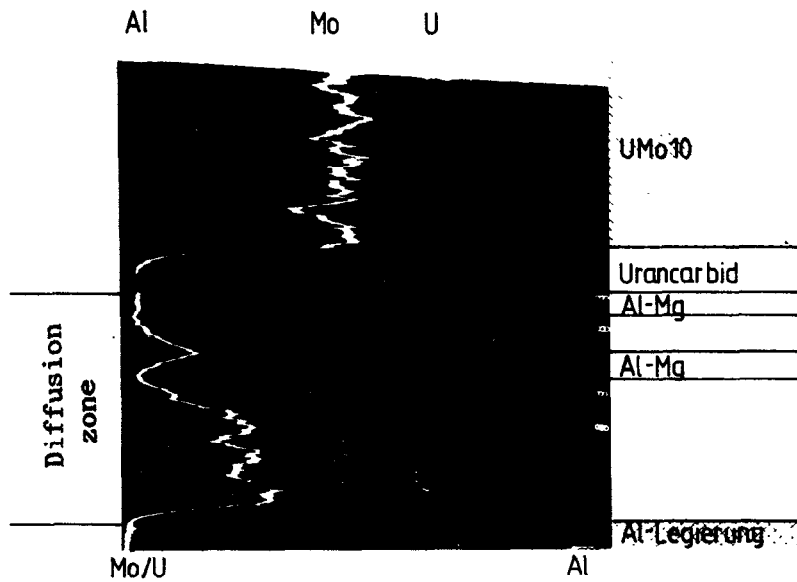


Fig. 7: MPMA linescans of the HIP-interface UMo10/AlMg3. HIP temperature: 550 °C (2 h) (900x).

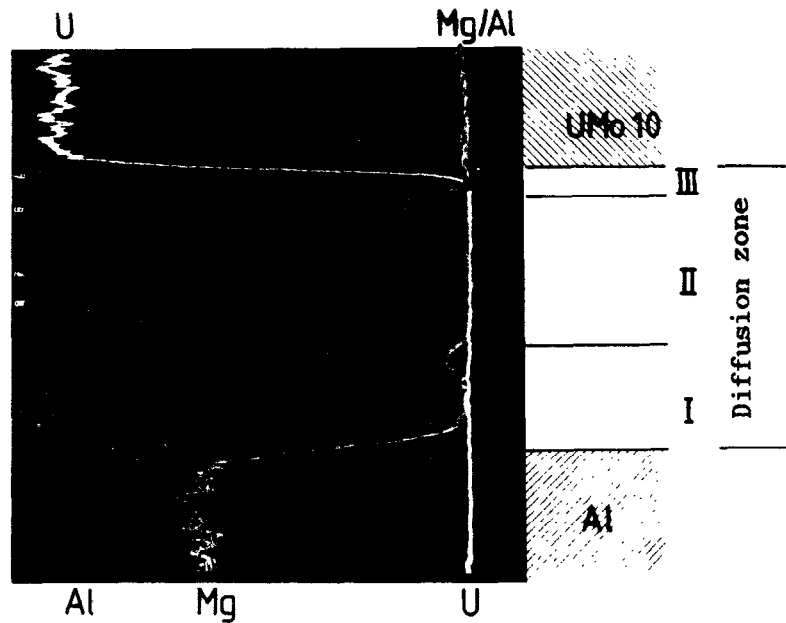


Fig. 8: MPMA linescans of the HIP interface UMo10/Ni/AlMg3. HIP temperature: 550 °C (2 h). Initial Ni layer thickness: 30 μm (900x)

As some of the intermetallic phases are expected to be brittle, cracks may develop during target operation, reducing the thermal conductivity at the interface and the mechanical stability of the cladding. Thus, target elements produced by hot isostatic pressing of UMolO and Zry-2 without Ni-bonding are expected to perform best.

This assumption is backed by tensile and shearing strength measurements of the UMolO/cladding interfaces. An example is given in Fig. 9, showing the highest shearing strength for the UMolO/Zry-2 interface and a HIP temperature of 800 - 900 °C. In agreement with tensile test data, the shearing strength is lower for the UMolO/Ni/Zry-2 version and the target elements with aluminium-based claddings independent of the presence of a Ni-bonding. The increase in strength with HIP temperature holding time of the UMolO/Ni/Zry-2 interface is due to the reduction of the Ni-bonding layer thickness during hot isostatic pressing.

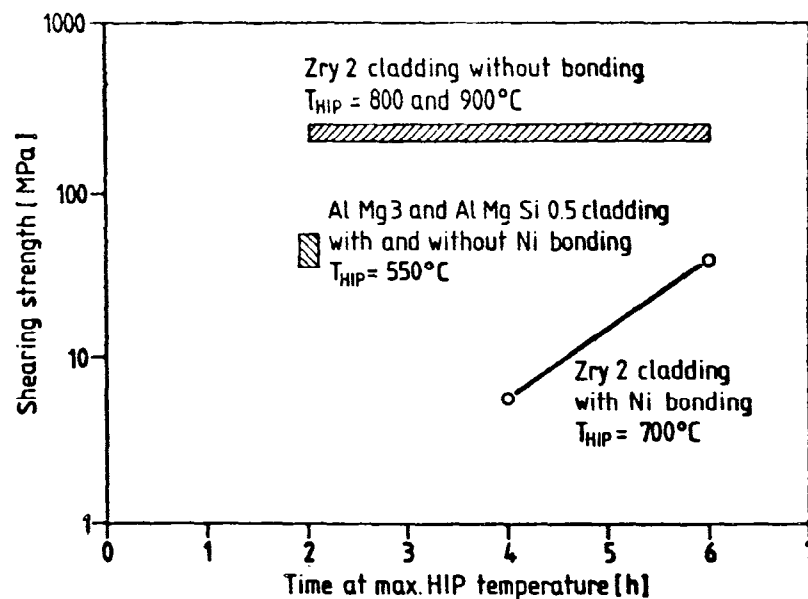


Fig. 9: Shearing strength of different UMolO/(bonding)/cladding interfaces as a function of HIP temperature holding time