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ADVANCES AND PERSPECTIVES IN U-MO MONOLITHIC AND DISPERSED FUELS.

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ABSTRACT

Development advances in uranium-molybdenum monolithic and dispersed fuels are presented so as to look forward additional alternatives to cover HEU-LEU conversion possibilities.

Preliminary results on post-irradiation examination of one zircaloy-4 cladded U-7Mo monolithic 20% enriched miniplate irradiated during the RERTR-7A experiment shows good performance.

With this encouraging results, new developments in monolithic fuels are being designed.

Process capabilities and performance prediction in dispersed fuels are being tried with the following characteristics:

1) Aluminum matrix alloyed with germanium, instead of silicon.
2) Coated U-Mo particles with silicon, germanium and nickel, and aluminum alloyed with these elements, so as to reduce interaction kinetics.
3) Incorporation of porous alumina in the matrix, to adsorb gaseous fission products.

1. Introduction.

Two miniplates of monolithic LEU U-7Mo with zircaloy-4 cladding elaborated in CNEA were shipped to INL [1, 2], for irradiation in the ATR jointly with other 23 miniplates, during the RERTR-7A experiment.

Diffusion couples of U-7Mo/Zry-4, hot co-lamination of three superposed Zry-4 plates and several prototypes, as the one shown in figure 1 were elaborated, characterized and tested with depleted uranium to set up process variables and fabrication conditions.

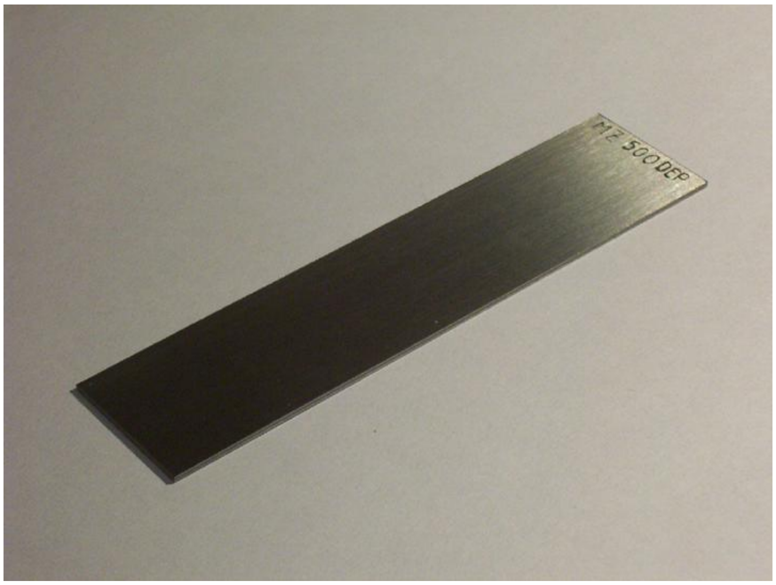

Figure 1. *Similar miniplate (100 x 25 x 1 mm^3) as those irradiated in experiment RERTR-7A at the ATR, INL.*

The characteristics of the irradiated plates are shown in table 1. They have a total thickness of 1 mm and meat thicknesses of 0.25 and 0.50 mm and they are identified as MZ25 and MZ50, respectively. The thicker meat miniplate had a cladding thickness of 0.25 mm.

Internal measurements were performed from X-Ray radiographies and uranium densities were calculated from geometrical dimensions and weights measured before final elaboration. Miniplates were also ultrasonically tested to check for 100% U-7Mo/Zry-4 bonding.

Table 1. *Geometrical and density characteristics of miniplates MZ25 and MZ50.*

Miniplate	MZ25	MZ50
Plate thickness (mm)	0.99	1.01
Meat thickness (mm)	0.26	0.51
Meat width (mm)	18.8	18.6
Meat longitude (mm)	73.0	71.0
Total U (g)	5.9	10.9
Meat density (gU/cm3)	16.5	16.2

The irradiation finished in March of this year and the as-run physics calculation obtained from the power history are synthesized in table 2. Total burn up, total density fissions and maximum heat fluxes are indicated for both irradiated miniplates.

Table 2. *As-run physics calculations.*

Miniplate	**Burn-up**	**Fission density**	**Max. heat flux**
ID	%	f/cm^3	W/cm^2
MZ25	38	2.7×10^{21}	135
MZ50	33	2.3×10^{21}	217

Post irradiation examination is already being performed of miniplate MZ25 at the Hot Fuel Examination Facility (HFEF) of the Material and Fuel Complex site of INL. Gamma scanning, dimensional measurements, and cuttings for chemical burn-up and metallographic inspection have already been performed (figures 2, 3 and 4). Table 3 show the total swelling of the miniplate and the dielectric layer measured by eddy currents. The dielectric layer corresponds to oxide formation and eventually crud deposits.

Table 3. *As measured mean values of thickness increment and fundamentally oxide layer.*

Miniplate	**Total swelling**	**Dielectric layer**
ID	%	microns
MZ25	3.6	2.6

The general appearance of the miniplate was that of a slightly blackened metallic surface.

2. Cladding thickness.

In the design of the U-7Mo monolithic plates a particular emphasis was given in thinning the usual cladding thickness of miniplates. This is evidenced in the 0.25 mm cladding thickness of miniplate MZ50, when the usual values of minimum thicknesses are around 0.38 mm. Fundamentally two reasons were taken in account. In first place the similar yield strengths of both materials and the fact that both materials before rolling had a planar shape allowed thinner claddings to be used. In second place, exploring the fabrication capability of doing thinner claddings allows an additional variable in the design of the conversion of high flux reactors, and specially of single fuel reactors.

This second argument was also recently showed by calculations of conversion of the FRM-II[3] where, using a smaller cladding thickness allows using a 28% enriched U-Mo fuel instead of a 33% enriched fuel. The total uranium inventory and number of plates were the same; hitherto, thinning the cladding implied increasing the water channel dimension.

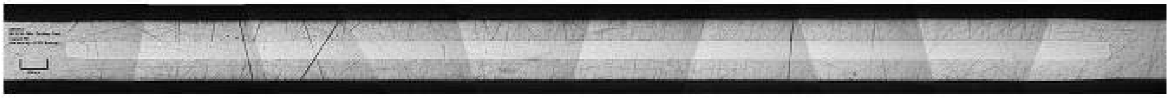

Figure 2. *Polished transverse cut of irradiated MZ25 miniplate.*

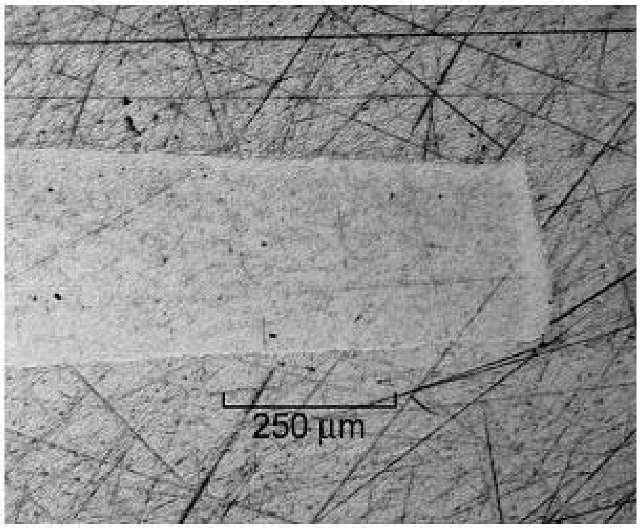

Figure 3. *Extreme side of U-7Mo monolithic meta surrounded by Zr-4 clad.*

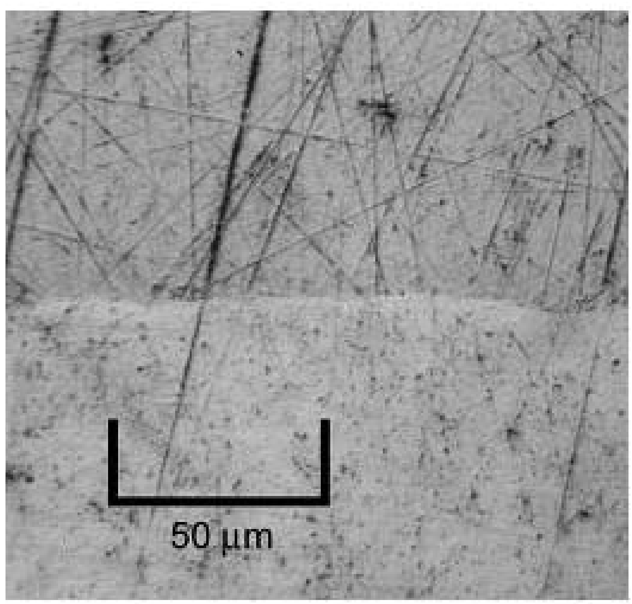

***Figure 4**. High magnification, interphase between U-7Mo meta (bottom) and Zr-4 clad (top).*

It is illustrative to show how the quantity of neutrons produced in each 235U fission depends on the moderation ratio for different enrichments. A simple calculation of k_∞ (k infinity) can be performed for the case of homogeneous water reactors with different enrichments. The moderation ratio can be expressed as the ratio between the hydrogen atoms and the 235U atoms. This dependence is shown in figure 5, where calculations were performed for the cases of 93 %, 28 % and 20 % enriched uranium. The 93% enrichment was calculated for a U3Si2 fuel and the smaller enrichments correspond to U-7Mo fuels. The moderator was light water.

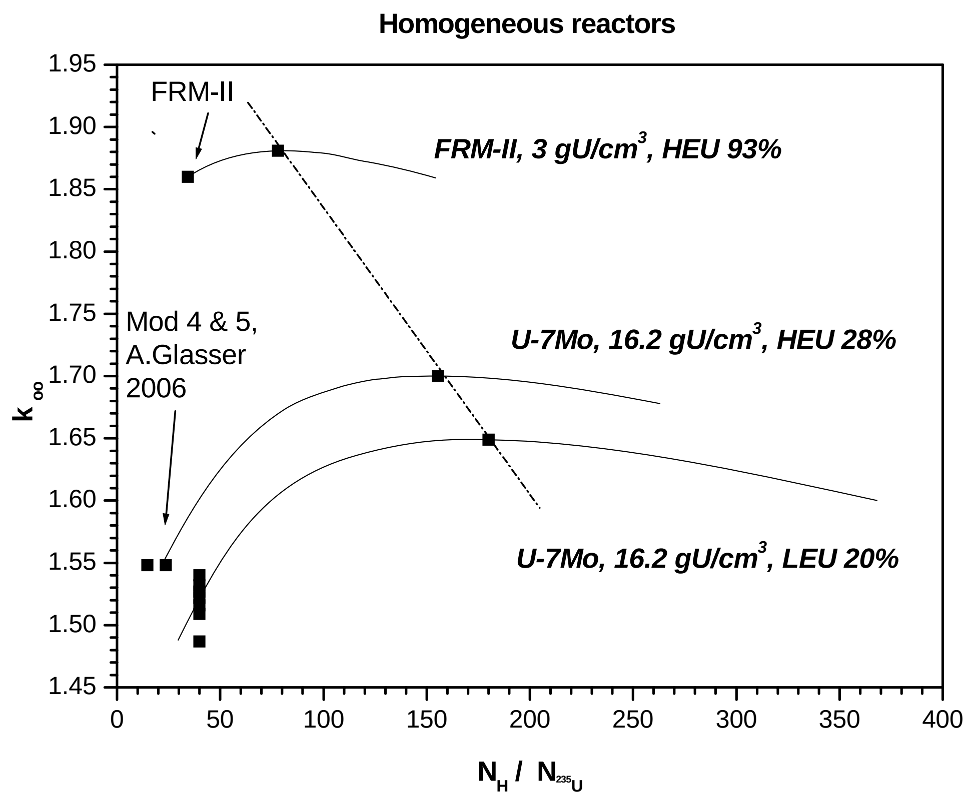

Figure 5. *Dependence of k_∞ on the moderation ratio for different uranium enrichments.*

From figure 5 it is seen that as enrichment diminishes, the maximum k_∞ correspond to higher values of moderation. The direct explanation of this effect is the resonant absorption of neutrons at intermediate energies ought to the higher abundance of 238U as enrichment decreases. In the 93% enrichment curve it is indicated explicitly the value of k_∞ corresponding to the FRM-II moderation relation that is 34. The relation is such that the reactor is sub-moderated and has a negative temperature coefficient. Points obtained by Glasser calculations [3], are identified also in figure 5 for cases corresponding to 28% enriched uranium.

The aligned points in figure 5, corresponding to a moderation ratio of 40, are expanded in figure 6 for a planar periodicity of plates and water channel similar as that in the FRM-II fuel. It shows the dependence of k_∞ with the meat thickness, for a fixed Zry-4 cladding thickness of 0.15 mm and water channel dimensions that varies between 1 to 5 mm. Obviously the number of plates should change and are indicated in numbers in the curve of figure 6.

It is clear from figure 5 that higher moderation ratios are desired in the design of LEU fuels. From figure 6 it is seen that auto-shielding effects are not evident in ranges where usual design periodicities between plate and water thicknesses are normally used.

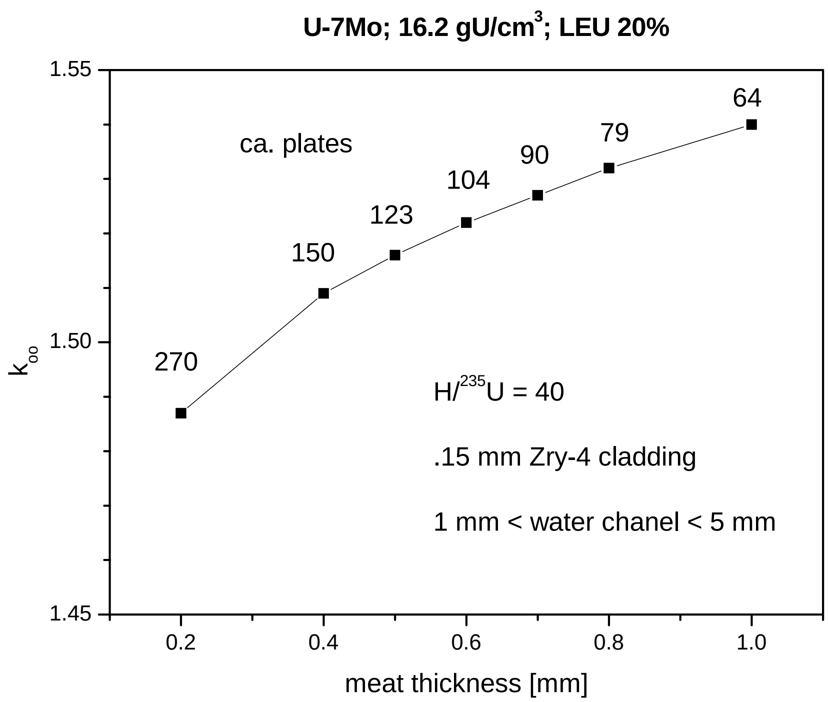

Figure 6. *Dependence of k_∞ with meat thickness for a constant moderation ratio $H/^{235}U = 40$.*

The reduction of cladding thickness is one of the main variables that can be managed in the conversion of HEU fuels to LEU releasing space for increasing moderation. It can easily be shown that with a 0.15 mm cladding thickness a better moderator/fuel relation can be optimized obtaining an equivalent k_{ef} . The relation between total cladding thickness and meat thicknesses, that is usually greater than 1 (~ 1.27) for plate type fuels, can be reduced to values smaller than 1 (~ 0.5), increasing the neutron moderation. Technologically, cladding thicknesses can only be reduced using stainless steel or zirconium alloys in the case of monolithic fuels.

Also, the moderator can be introduced in the meat itself, with the possibility of incorporating of zirconium hydride (ZrH_2) as a moderator in the meat, with a uranium density higher than 10 gU/cm^3. The introduction of ZrH_2 in the meat can probably be prepared by using powder metallurgy methods which are being tested. This can introduce the capability of producing monolithic fuels with powder metallurgy methods that can also have some advantages in the production of heterogeneous and asymmetric meats and reducing scrap and/or re-melting operations.

3. Interaction UMo/Al.

Undesired porosity in the aluminum side of the interaction zone between U-Mo and Al that was detected in dispersed fuels is now being reported to be present in irradiated monolithic fuels [4] (RERTR-6) with aluminum cladding. One possibility to reduce this undesired interaction is the incorporation of a diffusion barrier in between cladding and meat. The material to be chosen as coating should not interfere and, if possible, help in the selected bonding process.

Although some evidence has been observed so as to eliminate this problem in dispersed fuels with the incorporation of silicon to the aluminum matrix, work is being continued on several different alternatives, such that some of them can be extrapolated to be also used in monolithic fuels or simply can be complementary solutions.

3.1. Alloying the aluminum matrix.

Germanium, as silicon and tin are from the same group in the periodic table of the elements and has been early shown that inhibits the formation of UAl_4 and hitherto stabilizes the compound UAl_3. It is expected that can have a similar effect on reducing undesired porosity in dispersed fuels. One reason to be explored for its use, as germanium does not form so many complexes as silicon, is simplifying back end options.

A 420° C eutectic is present in the Al-Ge phase diagram and special precautions should be attained in the fabrication process such that this temperature is not over passed. The maximum Ge atomic solubility in Al is slightly higher than Si and the U-Ge phase diagram has similar features as the U-Si phase diagram.

3.2. Diffusion barriers.

Previous results in Si coating techniques showed contaminated deposits, probably with precipitates of mixed oxides, and fractured deposits when the thickness was in the order of one micron. Improvements are being implemented using H_2 atmosphere, such that reduction of U-Mo oxides can take place. Also, germanium can be deposited using similar techniques as with silicon.

From the point of view of avoiding brittle coatings it is important to try to use ductile materials for the coating of U-Mo alloys. Nickel and niobium have been used in coating processes of uranium compounds, and at first sight, nickel can probably work well as a diffusion barrier. Niobium is best but presents more problems for implementing a coating process.

Diferent deposition techniques have been and are being tried: chemical vapor deposition (CVD), liquid immersion, high temperature solid diffusion and electro less in the case of nickel. Fuel particles and foils are being coated.

Coatings with aluminum alloys of Si and Ge are still being tried and some expieriences will be done trying to obtain a coating of the very stable cubic intermetallic AlNi.

Coatings should also protect U-Mo particles since, if they are exposed to air, the continuous oxidation lowers the density of the material. After one year of fabrication, HMD particles have diminished its density in a value lightly smaller than 1 %.

3.3. Dispersed porous alumina.

In a dispersed U-7Mo meat of 8 gU/cm3 with a 80 microns size fuel particles and a 50% burn-up, an equivalent of approximately twice the meat volume of fission gas products -at normal temperature and pressure (NTP) conditions- are released into the Al matrix. These fission gases will diffuse threw Al grain boundaries and can nucleate and grow at heterogeneous sites. These fission gas products, xenon and krypton, are the driving forces that generate an excess pressure and consequently undesired porosity in the aluminum side of the interaction zone with U-Mo.

If these fission gas products could be adsorbed in a high specific surface material, pressure would be smaller, and the swelling driving force also. One material that is being tested is porous alumina so as to know the equilibrium pressures at a fuel plate working temperature as a function of adsorbed gases. Porous alumina can be incorporated as dispersed powder in the aluminum matrix of a dispersed fuel, and to avoid process problems it should not occupy more than 10% of the meat volume.

Since having the porous alumina activated inside the pressed compact is difficult to achieve, it was first tested the pressure conditions of U-7Mo powder in a closed chamber, simulating the inside pressure and temperature conditions of a hot rolled picture and frame ensemble.

Figures 7 and 8 show the pressure evolution from an initial value of 800 mbar air after a peak temperature of 400/500 °C in less than one hour. The volume of the chamber was of approximately 3 liters and runs with 64 and 130 grams of U-7Mo HMD powder were performed. The first run was the one with a sample of 64 g, the second run was with a new sample of 130 g, and the third run used the same sample as before. The initial increase in pressure corresponds to adsorbed water and quickly all of the three performed runs achieved a final pressure of 200 mbar. The third run had a slower kinetics than the fresh samples, but the final pressure was the same. Clearly, the U-7Mo powder is incorporating oxygen and nitrogen. Surely this will be the initial pressure conditions inside a picture and frame ensemble before hot rolling.

At pressures around 200 mbar no relevant incorporation of air was noticed using only powdered porous alumina. New runs are being planned at higher pressures using an inert gas to obtain the equilibrium pressures at the working temperatures of a fuel plate

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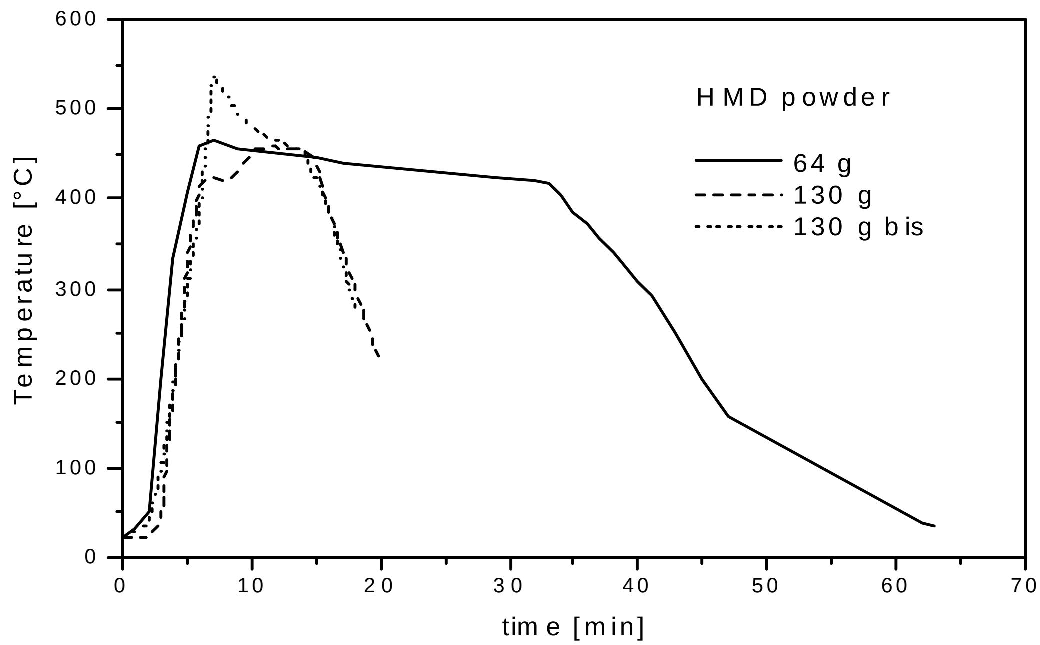

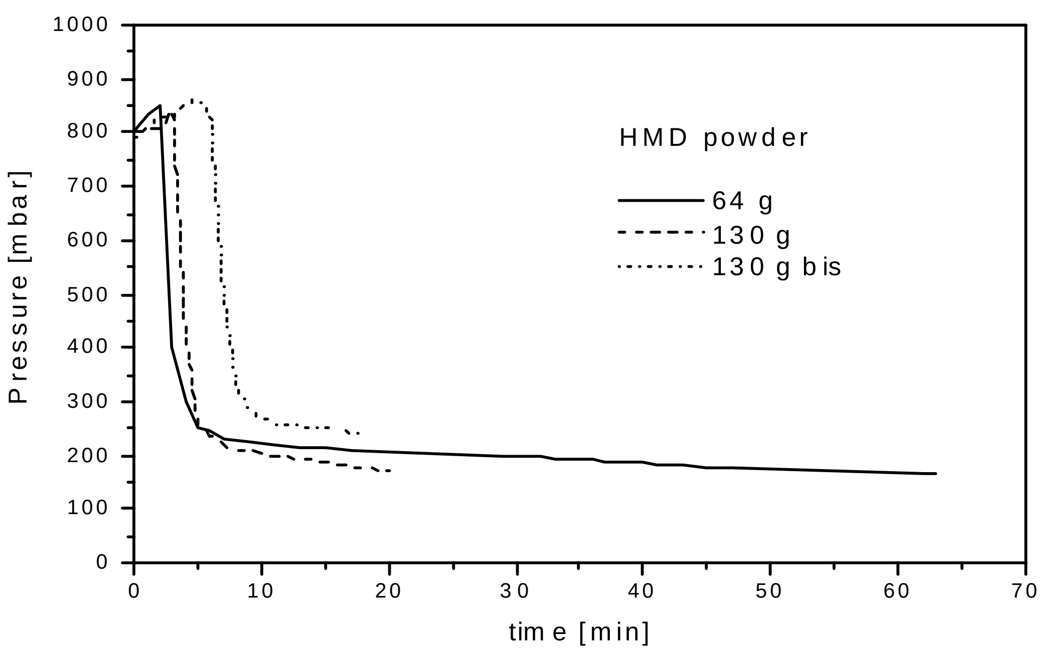

Figures 7 and 8. *Temperature and pressure evolution of U-7Mo powder exposed to an air atmosphere in a closed chamber.*

4. Conclusions.

4.1. Monolithic U-7Mo with Zry-4 cladding

Preliminary results of PIE of monolithic U-7Mo with zircaloy-4 cladding show promising behavior since it had been tested only up to 38% burn-up. Higher fission density is needed to cover higher burn-ups.

It is important to emphasize the ease of hot rolling fabrication -that has to be scaled up to full size plates- and the gentle interaction zone, that it is not expected to present problems at higher burn-ups.

The possibility of using smaller cladding thicknesses when zircaloy is emplyed allow design improvements in LEU conversions and reactor design

4.2. U-Mo interactions with Al

Coating processes of U-Mo particles and foils are being improved using reducing atmospheres.

Incorporation of fixed porosity (porous alumina) in the Al matrix is being studied and tested as plenums to adsorb fission gas products, as a mean to reduce the effective pressure at preferential localized sites.

5. Acknowledgments.

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6. References.

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4. R. Finlay, personal communication and this RERTR-2006 meeting.