DEVELOPMENT OF UZr ALLOY FOR THE TRIGA/IPR-R1 REACTOR FUEL

[1,2]Silva, E. G.; [1]Cardoso, P. E.; [1]Ferraz, W. B.

[1]Centro de Desenvolvimento da Tecnologia Nuclear - CDTN/CNEN
Rua Professor Mário Werneck, s/ n^0 – 31123-970
Belo Horizonte – MG - BRAZIL

[2]Departamento de Engenharia Nuclear - CCTN
Universidade Federal de Minas Gerais – UFMG
Belo Horizonte – MG - BRAZIL

Presented at:
The 1998 International
Reduced Enrichment for Test Reactor Conference
Sao Paulo, Brazil
October 18-23, 1998

DEVELOPMENT OF UZr ALLOY FOR THE TRIGA/IPR-R1 REACTOR FUEL

[1,2]Silva, E. G.; [1]Cardoso, P. E.; [1]Ferraz, W. B.

[1]Centro de Desenvolvimento da Tecnologia Nuclear - CDTN/CNEN
Rua Professor Mário Werneck, s/ n^0 – 31123-970
Belo Horizonte – MG - BRAZIL

[2]Departamento de Engenharia Nuclear - CCTN
Universidade Federal de Minas Gerais – UFMG
Belo Horizonte – MG - BRAZIL

ABSTRACT

This paper reports the fabrication development at CDTN of the UZr alloy for the TRIGA/IPR-R1 reactor fuel. A comparative study of the melting of UZr alloy by using vacuum consumable-electrode arc (VAR) and vacuum induction melting (VIM) processes is described. The characterization of the alloy was done by chemical, diffractometric, metallographic, delayed neutrons and mechanical testing. Both processes have shown advantages and disadvantages. In the case of VAR process, it was necessary to remelt the ingot to homogenize the alloy. The influence of the observed contamination by C in the VIM process on the alloy neutronic and mechanical properties is a case for further studies.

1. INTRODUCTION

A power increase of the CDTN/CNEN TRIGA/IPR-R1 reactor from 100 KW to 250 KW is foreseen for the near future. To meet the increase of U_8ZrH_n fuel demand, the CDTN/CNEN Materials and Nuclear Fuel Department, in the scope of the materials and nuclear fuel research program, is carrying out a project aiming at the technology development of the production of (U_8ZrH_n) fuel. This development is performed in two stages. The first one, where this work is inserted, consists in accomplishing a comparative study for obtaining UZr alloy using two melting processes, namely, one in arc furnace with consumable electrode (VAR), and the other one in induction furnace (VIM), both being operated under vacuum. In the next step, the alloy hydriding process is intended.

In the VAR process, the zirconium sponge is mechanically mixed with pieces of natural uranium and pressed in cylindrical samples. These samples are TIG (Tungsten Inert Gas) welded, and the obtained UZr electrodes are melted in the arc furnace. After the melting, the ingot is hot forged (Swaging) and finally machined in a lathe. It should be pointed out that the UZr alloy production technology was originally developed by General Atomic Co [01].

In the VIM process, the pressing and welding steps are eliminated. The zirconium sponge and natural uranium pieces are directly melted in a graphite crucible without internal coating,

and poured out in a copper ingot mold. As uranium and zirconium are highly reactive, the contact of these metals in liquid phase with the graphite crucible presents the carbon contamination inconvenience. Therefore, this production route is made possible decreasing melting effervescence time and consequently the carbon contamination. Then, a comparative study of these two technologies is made.

The alloy characterization was performed through chemical, delayed neutrons, diffractometric, metallographic analyses, and mechanical tests to evaluate the feasibility of both processes.

2. EXPERIMENTAL PROCEDURE

All VAR technology for UZr alloy concerning equipment, components, and tools was developed in CDTN [02]. In parallel, the technology of uranium and zirconium pressing, electroctrode TIG welding and the ingot mechanical diameter reduction by rotary swaging were also worked. The load pressing was established at 7.5 ton/cm^2, in order to obtain rods with a 150 mm length and 24 mm diameter, with a density reaching 78% of the theoretical density. The TIG process in sealed chamber was used in rod welding. After welded to each other, the rods originated an electrode with a 450 mm length and a 24 mm diameter. The melting parameters were 850 A, with 29 V, resulting in a melting rate of 0.24 Kg/min. The obtained ingot had a 50 mm diameter and a 75 mm length. The mechanical forming was accomplished in rotary swaging and the ingot diameter was reduced to 24 mm after 6 reduction steps. Between each reduction step, it was necessary to proceed a heat treatment at 800^{o}C, during 5 minutes, in order to eliminate the work hardening effects. After the last reduction step, the rod was hot pressed in a small manual press, to eliminate any resulting warping from the forging. The vacuum induction melting was used to evaluate the VIM process. As this process eliminates the pressing and welding steps of the electrode, a better distribution of the uranium along the ingot longitudinal and cross section directions is expected. Zirconium, with a higher melting point and a lower relative density than uranium, was loaded in the bottom of the graphite crucible, in order to benefit from the highest temperature at the crucible low position. Thus, the uranium melting was delayed, consequently reducing its contact time with the carbon, which presents a high solubility in liquid uranium [03,04]. The aim of the first experiences was to adjust the melting in the shortest time, so avoiding the alloy solidification inside of the crucible during the casting process and allowing the bathing a larger effervescence (so eliminating the volatile impurities) and also the segregation of the non volatile impurities to the surface [05].

3. RESULTS AND DISCUSSION

3.1 U-Zr Alloy Obtained in Vacuum Arc Furnace

3.1.1 Chemistry (CA) and Delayed Neutrons (DNA) Analyses

The results of chemistry (CA) and delayed neutrons (DNA) analyses are shown in Table 1.

Table 1 -Zirconium and Uranium Content Along the Ingot ($^w/_o$)

ZIRCONIUM			URANIUM (CA)			URANIUM (DNA)		
A	B	C	A	B	C	A	B	C
94	93	93	5.0	6.3	5.6	5.5	6.6	6.1

These results show that a significant variation in the distribution of uranium along the ingot occurred. As suggested by W.W.DYRKACZ [06] during the pressing step, there was a poor distribution of the components along the electrode. It was also noticed that the uranium content determined by chemical and delayed neutrons analyses presented a variation of about 10%. To attenuate this misdistribution and confirm it as foreseen by General Atomic [01], this ingot was remelted. A satisfactory homogeneity was obtained as shown in Table 2.

Table 2-Distribution of Uranium After Remelting in Arc Furnace ($^{w}/_{o}$)

URANIUM (DNA)		
A	B	C
5.4	5.9	5.5

3.1.2 - Diffratometric Analysis

The diffratometric analysis (Figure 1) has revealed a predominance of the phase α(Zr) and a small amount of the phase $\delta(UZr_2)$, as expected by the UZr alloy phase diagram [01-04].

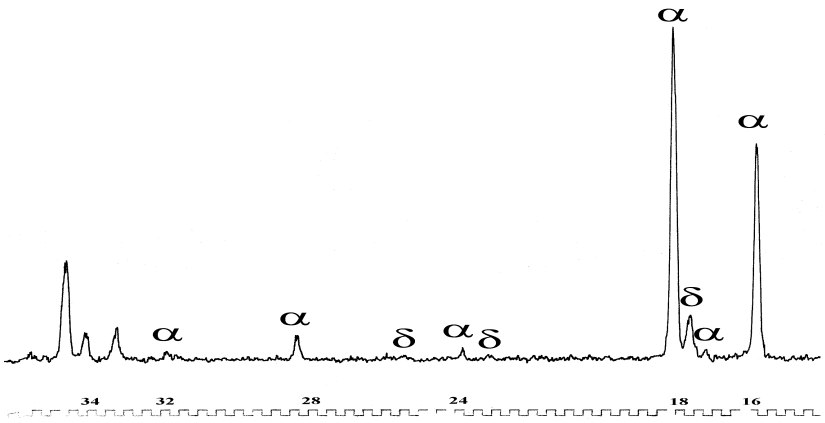

Fig. 1 – Diffratometric Analysis of the UZr Alloy in VAR

3.1.3 -Metallographic Analysis

Figure 2 shows the metallographic analysis of an ingot in rough-cast, and Figure 3 shows the sample after annealing at 900^{o}C for 5 minutes.

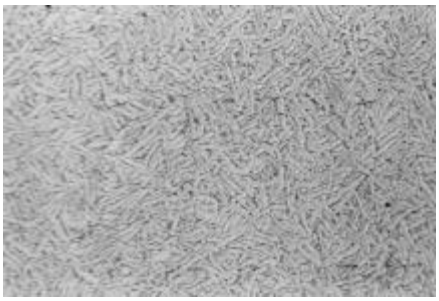

Fig. 2 – Sample in Rough-cast – 500X

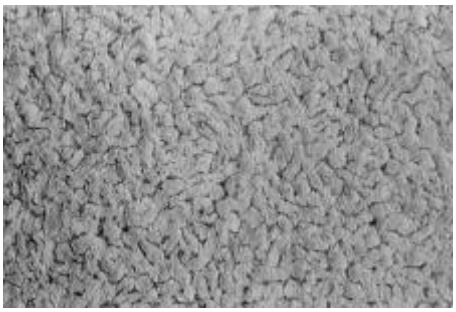

Fig. 3 – Sample After Annealing – 500X

As one can see from Figure 2, phase α(Zr) is predominant (clear tonality), whereas phase $\delta(UZr_2)$ exists in a smaller amount (dark tonality). The presence of inclusions is also noticed, probably originated from the impurities present in the raw materials. In Figure 3 is shown a change from acicular (rough-cast) to equiaxial grain structure after annealing.

3.1.4 Mechanical Tests

- Tensile strength - σ_μ 75.6 kgf/mm^2 ≅107549 Psi.
- Yield strength - $\sigma_\gamma(0,2)$ ≅ 68.9 kgf/mm^2 ≅ 97975 Psi.
- Modulus of elasticity-E≅ 1.8x10^3 kgf/mm^2 ≅ 2.6x10^6 Psi
- Strain - e= ΔL/Lo ≅ 2.9 mm/32mm ≅ 9.0%

Brinell hardness (HB) tests were made in the traverse section of the samples used in the tension test. The mean value obtained from a set of five evaluations was 218 HB. The tension and Brinell hardness tests were carried out in the ingot after na annealing at 900^{0}C for 5 minutes.

3.2 U-Zr Alloy Obtained in Vacuum Induction Furnace

3.2.1 Chemistry (CA) and Delayed Neutrons (DNA) Analyses

As it can be observed from Table 3, the ingots presented an uranium homogeneous distribution, so confirming W.W.DYRKACZ [06]. This homogeneity has also occured in ingot FI-04, which only spent 0.25 minute in effervescence after its total melting. Note that this time was of 1 minute for all other ingots.

Table 3 - Chemistry and Delayed Neutrons Analyses ($^w/_o$)

INGOTS	ZIRCONIUM				URANIUM (CA)				URANIUM (DNA)			
	A	B	C	D	A	B	C	D	A	B	C	D
FI-01	91	91	92	91	7.9	7.9	7.9	7.9	8.1	7.8	7.9	7.7
FI-02	93	92	90	92	7.9	7.9	7.9	7.8	8.1	7.9	7.9	8.0
FI-03	91	91	91	92	7.8	7.8	7.8	7.8	7.8	7.8	7.9	8.0
FI-04	92	92	91	91	-	-	-	-	8.1	8.1	8.3	8.2

Tables 4 and 5 present carbon content, before and after melting in induction furnace, respectively.

Table 4 - Carbon Content Before Melting in Induction Furnace ($^{w}/_{o}$)

CHARGE	CARBON
Zr	0.010 a 0.020
U	0.083

Table 5 - Carbon Content After Melting in Induction Furnace ($^{w}/_{o}$)

INGOTS	CARBON			
	A	B	C	D
FI-01	0.32	0.31	0.32	0.32
FI-02	0.29	0.30	0.29	0.29
FI-03	0.30	0.28	0.32	0.29
FI-04	0.20	0.19	-	0.20

A very significant level of carbon contamination in the ingots FI-01, FI-02 and FI-03 was verified. It should be noticed, however, that ingot FI-04 showed a smaller contamination, because the alloy was given only a short contact time with the graphite crucible.

It was made an attempt to combine VIM and VAR processes. It was expected that the oxygen present in the arc furnace chamber and the electric arc high temperatures could promote the reaction of oxygen with carbon producing CO, thus having C content in the ingot decreased. Through this, pure zirconium melting was accomplished in the induction furnace and one of the ingots was remelted twice in the arc furnace. Results are shown in Table 6.

Table 6- Pure Zirconium Carbon Content Before and After Remelting ($^{w}/_{o}$)

PURE ZIRCONIUM	CARBON
After melting (VIM)	0.24 to 0.38
After first remelting (VAR)	0.14
After second remelting (VAR)	0.13

These results indicate that a VAR remelting results in significant carbon decrease. Then, expecting that this C decrease could also happen with the UZr alloy, ingots FI-01/FI-02/FI-03 were remelted in arc furnace. Results can be seen in Table 7.

Table 7 -Carbon Content After Remelting in Arc Furnace ($^{w}/_{o}$)

INGOTS	CARBON
FI-01	0.32
FI-02	0.29
FI-03	0.29

The expected carbon content decrease in the UZr alloy did not occur, as shown above. It seems that the C decrease, which happened with pure zirconium, was due to the dissolved carbon, and not to that percentage forming the carbide phase as seemed to occur with UZr alloy. After hydriding, the alloy may present the following composition (atomic percent):

- U = 1.6
- Zr = 48.9
- H = 48.9
- C = 0.6

One can thus obtain a quaternary alloy, in which we will have the addition of 0.6% in carbon atoms, in respect with the original alloy (U_8ZrH_n). Although references that link the influence of carbon in the general properties of this fuel have not been found, it should be pointed out that the presence of carbon in this alloy can modify its nuclear properties, due to an existing similarity between carbon and hydrogen, as to neutronic moderation. The C contamination can also decrease the corrosion resistance, as mentioned by LUSTMAN & BENJAMIN [08], due to the carbide phase preferential corrosion.

3.2.2 - Diffractometric Analysis

The diffractometric analysis (Figure 4) showed a predominance of the α phase (Zr), followed by a smaller amount in phase δ (UZr_2) and traces of zirconium carbide (ZrC).

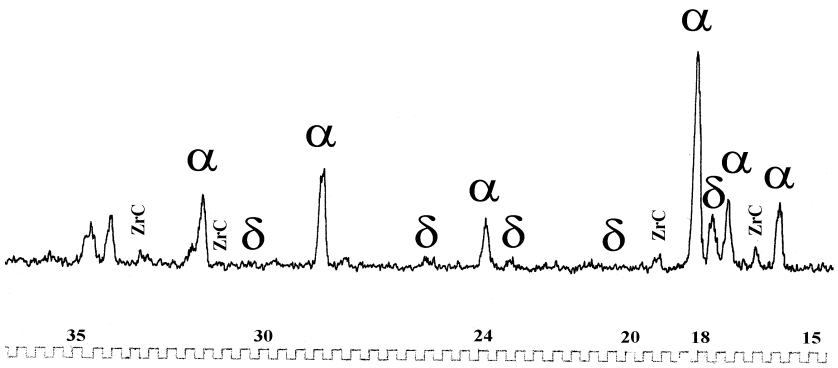

Fig. 4 – Diffratometric Analysis of the UZr Alloy in VIM

3.2.3 –Metallographic Analysis

The metalographic analysis was accomplished in rough-cast samples, after annealing at 900^0C for 5 minutes.

Figure 5 shows sample structure in the rough-cast step. There is a predominance of the α phase (Zr) in the clear tonality and δ phase (UZr_2) appears in a smaller amount in the dark tonality. The presence of aligned inclusions, probably rich in carbon, can be also verified. It becomes evident from Figure 6 that the structure was obtained after the annealing. Morphology here changes from acicular to equiaxial.

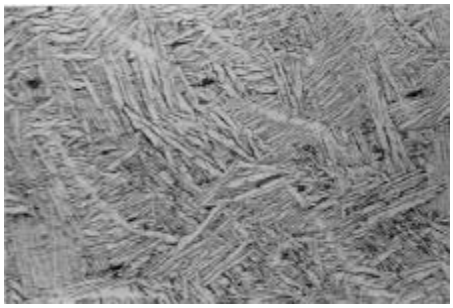

Fig. 5 – Sample in Rough-cast – 500X

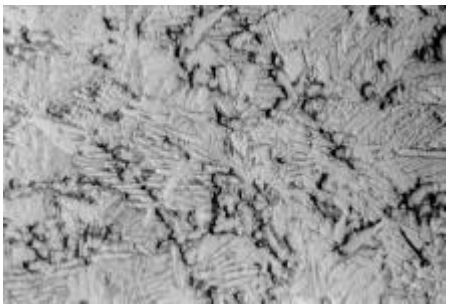

Fig. 6 – Sample After Annealing – 500X

3.2.4 Mechanical Tests

- Tensile strength - $\sigma_\mu \cong 84.8$ kgf/mm^2 $\cong$120585 Psi.
- Yield strength - $\sigma_\gamma(0.2) \cong 74.3$ kgf/mm^2 $\cong$ 105654 Psi.
- Modulus of elasticity-E$\cong 2.3 \times 10^3$ kgf/mm^2 $\cong 3.3 \times 10^6$ Psi
- Strain - e= $\Delta L/Lo \cong 2.0$mm/32mm $\cong$ 6.3 %

Brinell hardness mean value for a set of five evaluations was 247 HB. Tension and Brinell hardness tests were accomplished in an ingot submitted to annealing at 900^0C for 5 minutes. The tensile strength for the arc furnace ingot was $\sigma_\mu = 75.6$ kgf/mm^2 $\cong$ 107549 Psi, which is 7% above the value found by General Atomic [08]. However, the tensile strength found for the induction furnace ingot ($\sigma_\mu \cong 84.8$ kgf/mm^2 $\cong$ 120585 Psi) was approximately 20% above that found by General Atomic. Such a sharp increase in tensile strength for the induction furnace ingot, according to LUSTMAN,BENJAMIN[09], is associated with the presence of extremely hard carbides. Strains of 9.0 % in the arc furnace ingot and of 6.3 % in the induction furnace ingot were also verified. Note that the 247 HB hardness, obtained for the induction furnace ingot, compared with the 218 HB value for the arc furnace ingot, is in agreement with the increment in yield strength. As foreseen by LUSTMAN, BENJAMIN [09], the formation of zirconium carbides significantly influences zirconium mechanical properties, as well as the properties of its alloys, mainly hardness.

4. CONCLUSIONS

1) The VAR process has proved appropriate for the UZr alloy obtention. However, adapting the uranium distribution in zirconium during the electrode press step shall be necessary, as well as remelting the ingot.

2) The UZr alloy, obtained by VAR process, presents mechanical properties similar to those depicted in literature;

3) With respect to the VAR process, the VIM process presented here offers operational advantages, because preparing the electrode for the pressing and welding is not necessary.

4) One of the advantages of the VIM process is the alloy homogenization with only one melting; its main disadvantage, however, is the alloy contamination by carbon;

5) Alloy contamination in the induction furnace process can be minimized, if we reduce to 0,25 minutes the alloy effervescence time after the charge is totally melted.

6) Carbon contamination seems to be unavoidable in the VIM process. Present in small amounts, this carbon can compete with the zirconium hydride (ZrH_n) as moderator. Therefore, establishing the influence of this contamination in the fuel U_8ZrH_n is essential to obtain a reference parameter, mainly regarding its nuclear properties.

7) The presence of carbon in the UZr alloy induces changes in its mechanical properties.

5. REFERENCES

[01]-General Atomic, Technical foundations of TRIGA.San Diego: 1958, p.77 (GA-471).

[02]-BRAGA, DANIEL MARTINS; SILVA, EDUARDO GOMES; CARDOSO, PAULO EDSON; Estudo de Fabricação da Liga Uzr para Combustível de Reator Tipo TRIGA TRIGA, VI CGEN, tópico ciclo do combustível, 1996.

[03]-HARRINGTON,C.D. AND RUEHLE A. Uranium Production Technology, 1959.

[04]-WILKINSON,W.D.-Uranium Metallurgy Volume II: Uranium Corrosion and Alloys. 1th ed. New York:John Wiley & Sons, Inc., 1962-P.947-948.

[05]-FREITAS, C.T.DE; CINTRA, S.H.L.; GENTILE, E.F.; CAPOCCHI, J.D.T.-Fusão e Lingotagem de Urânio sob Vácuo em Forno Elétrico de Laboratório. Metalurgia, Vol.24, n125, p 279, Abril, 1968.

[06]-W.W.DYRKACZ, Metal Progress 75(5), 138 (1959).

[07]-LUSTMAN, BENJAMIN-Zirconium Production Methods. In: The Metallurgy of Zirconium. N.Y., McGraw-Hill Book Company.Inc., 1955. p 616-618.

[08]-General Atomic, Technical Foundations of TRIGA.San Diego:1958, p.69-72(GA471).

[09]-LUSTMAN, BENJAMIN -Zirconium Production Methods. In: The Metallurgy of Zirconium. N.Y. ,McGraw-Hill Book Company.Inc. , 1955.p.518-537.