PRESENT STATUS ON THE MECHANICAL CHARACTERIZATION OF ALUMINUM ALLOYS 5754-NET-O AND 6061-T6 IRRADIATED AT HIGH FLUENCES

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Abstract

The conception of the Jules Horowitz Reactor (JHR) requires to qualify at high neutron fluences the alloys which will be used for the tank, the core components and the experimental devices. To validate the choice of an aluminum alloy for the JHR tank, we started an extensive characterization program of Al-alloys irradiated in research reactors during at least 15 years.

For this program, we investigated some highly irradiated materials : the AG3-NET in annealed condition (5754-NET-O) and the 6061-T6. The 5754-NET-O is coming from components replaced during the refurbishment of the OSIRIS and ORPHEE reactors operated by CEA at Saclay (France). The components are the lattice structure of OSIRIS irradiated for 30 years (from 1966 to 1996), and the cold source shell of ORPHEE irradiated for 15 years (from 1980 to 1995).
The 6061-T6 alloy is extracted from rods used as beryllium plug during 32 years in the BR2 reactor located at Mol (Belgium).

This paper describes the mechanical tests (tension, fracture toughness) already performed on these materials. The effect of neutron fluence on mechanical properties of the AG3-NET-O (5754-NET-O) and 6061-T6 alloys is investigated.

Finally, an outline of the further program for the qualification of the JHR tank and experimental devices is provided for both alloys. In non-irradiated condition, erosion resistance tests are in progress, and welding ability is tested on different grades of 6061-T6. For the irradiated condition, an irradiation of specimens in the alloy chosen for the tank will be performed in OSIRIS. A great part of these specimens will be transferred into the JHR and used for a surveillance program of the tank.

1 - INTRODUCTION

The aluminum alloys of the 5000 and 6000 series have been extensively used in research reactors, because they have good mechanical properties at temperatures under 150°C, low neutron absorption and γ-heating, a reduced level of activation under neutron flux, and a good corrosion resistance in deionized water under 80°C.

The choice of an aluminum alloy allows to limit the operating temperatures of the JHR tank to a local maximum of 75°C in nominal conditions of neutron flux. To validate this choice for the JHR tank, we started an extensive characterization program of 5754-NET-O and 6061-T6 alloys irradiated in research reactors to thermal fluences of at least $5^{E}22$ n/cm^2.

Aluminum alloys of the 6000 series which are strengthened by a precipitation heat treatment are known to have a higher mechanical strength than in the annealed temper condition. Moreover, they have a relatively good mechanical stability under neutron flux [1, 2, 3, 4, 5] in contrast with alloys of the 5000 series, which are strongly hardened by the thermal neutron flux and become brittle at high fluences [6, 7]. This relative stability of their mechanical properties under neutron flux makes them attractive for use in research reactors where temperatures are low.

In France in the past, we extensively used the 5754-NET alloy in the annealed temper condition, where the NET specification corresponds to a modified chemical composition in order to reduce neutron absorption and activation, and to limit sensitivity to intergranular corrosion. Aluminum alloys of the 6000 series have not been used in the past in French research reactors, but 6061-T6 alloys were extensively used in American conception of core components. Therefore, we looked for highly irradiated components from 6061-T6 alloys available in foreign reactors. The BR2 reactor located at Mol (Belgium) having an American design is selected for the characterization of 6061-T6 material extracted from rods used as beryllium plug. This characterization is underway at SCK-CEN (Belgium).

The strengthening of aluminum alloys under neutron irradiation results from the joint effects of fast and thermal neutron flux :

The fast neutron flux creates point defects (interstitials and vacancies) and dislocations. The partial elimination of the interstitals on the dislocations and the grain boundaries causes an excess of vacancies which precipitate to form cavities observable at high fluences over $6^{E}22\ n_{fast}/cm^2$ [1].

The specificity of irradiated aluminum alloys is that aluminum transmutes under neutron flux to produce :

- helium by reaction (n,α) with fast neutrons
- hydrogen by reaction (n,p) with fast neutrons
- silicon by reaction (n,γ) with thermal neutrons producing ^{28}Al which desintegrate β^- into ^{28}Si.

In alloys of the 5000 series, the main alloying element Mg is in solution. The production of silicon under irradiation causes a precipitation of Mg_2Si, which induces an increase of mechanical strength and a loss of ductility.

In structural alloys of the 6000 series, the main alloying elements Mg and Si are fine precipitates obtained by heat treatment, therefore only the Mg content in excess of the stoechiometric composition Mg_2Si is available for further precipitation with silicon produced by irradiation. As precipitation of silicon alone is less efficient for strengthening, these alloys have a better stability under irradiation.

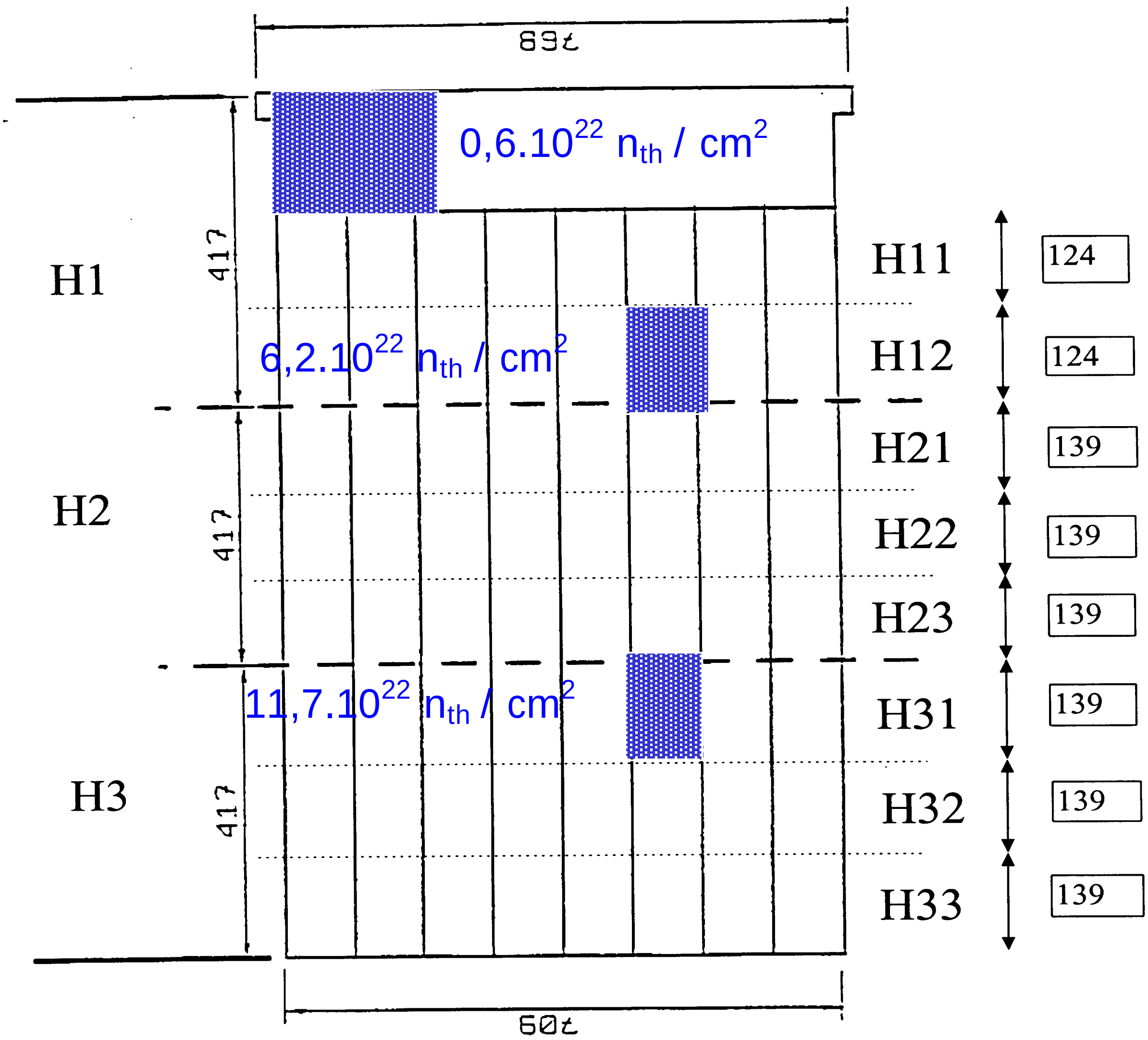
At high fluences, the strengthening mechanism by silicon precipitation prevails. Therefore, the mechanical characteristics of the alloys are generally plotted versus the thermal fluence.

2 - CHARACTERIZATION OF COMPONENTS IN AG3-NET-O

2.1 - AG3-NET-O irradiated with F_{th}/F_{fast} » 1

The core lattice structure of OSIRIS which was removed in 1996 after 30 years irradiation in light water at a mean temperature of 50°C, has been used for tensile tests.

Figure 1 : Core lattice structure of OSIRIS (dimensions in mm) irradiated with $F_{th}/F_{fast} \approx 1$



This structure in annealed AG3-NET is machined in a forged block and the cells have a wall thickness of 3 mm. The tensile specimens were cutt vertically at 3 different levels of one cell, chosen for his high neutron fluence (fig.1). In this cell, the ratio of the thermal neutron flux F_{th} (<0,625 eV) to the fast neutron flux F_{fast} (>0,9 MeV) is about 1.

As aluminum transmutes into silicon under thermal and epithermal neutron flux, the neutron fluence received by aluminum alloy components can be evaluated by measuring the silicon content. A correct estimation of the fluence requires to know the initial content in silicon before irradiation, and to perform neutron calculations taking into account the power history and the neutron spectrum of the reactor, and the position of the component.

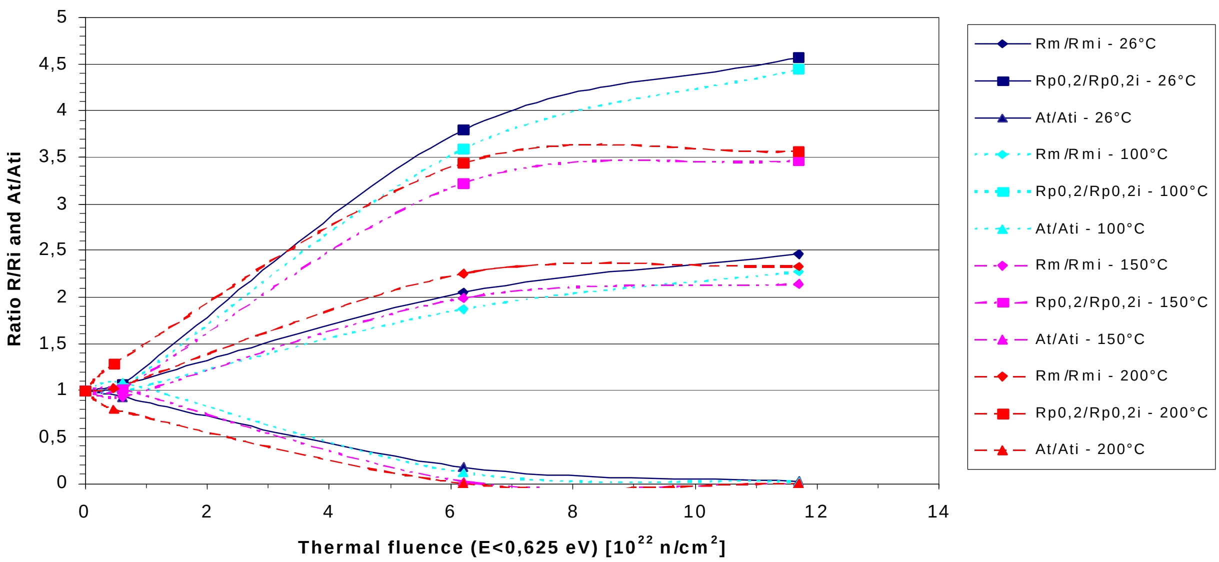
The silicon content was measured by optical emission spectroscopy of the vapor obtained by laser ablation of the sample : LA-OES technique [8]. For the neutron calculations, we used a simplified power history, assigning a mean power to each reactor cycle. The thermal neutron fluence (E< 0,625 eV) received at each level of the lattice structure was estimated with a precision of 14 to 18% ; the calculated values at the 3 levels are :

$F_1 = 6.1^E21\ n_{th}/cm^2$
$F_2 = 6.2^E22\ n_{th}/cm^2$
$F_3 = 11.7^E22\ n_{th}/cm^2$

At each level, specimens were tested at 26, 100, 150 and 200°C, and at each temperature, 3 specimens were tested [9]. The tests were performed with a static tensile device, with a strain rate of 4^E-4 s^{-1}, following the EN-NF 10002-1 standard.

At the lowest fluence F_1, the mechanical properties are in good agreement with those of the non-irradiated alloy : the alloy is practically not affected. At the highest fluences F_2 and F_3, the AG3-NET is severely hardened : at room temperature, the yield strength increases by a factor 4 to 4.5, whereas the tensile strength increases only by a factor 2 to 2.5, and the total elongation decreases drastically from about 23% to 4% at the middle fluence and under 1% at the highest fluence (fig.2).

Figure 2 : AG3-NET-O - F_{th}/F_{fast} =1
Evolution of yield, tensile strength and total elongation with fluence



At 150 and 200°C, the increase in strength reaches a limit at the middle fluence F2 over which it is constant ; at the same time, the total elongation drops to values under 1% at the middle fluence F2, and the alloy becomes brittle at higher fluences. This brittle behavior at high fluences is confirmed by SEM observations of the fracture surface.

2.2 - AG3-NET-O irradiated with Fth/Ffast » 100

The cold source shell SF1 of ORPHEE is an AG3-NET-O vertical tube (fig.3) made of a rolled sheet 6,3 mm thick, and TIG-welded over the whole length of the tube. This shell was irradiated in ORPHEE during 15 years from 1980 to 1995, in the heavy water reactor tank at a mean temperature of 40°C.

Tensile specimens were machined in the length of the tube at two vertical levels : one top level located outside the neutron flux and used as a non-irradiated reference, and one level situated in the maximum neutron flux, just over the cold neutron source. At each flux level, 3 tensile specimens were tested at 25°C [10].

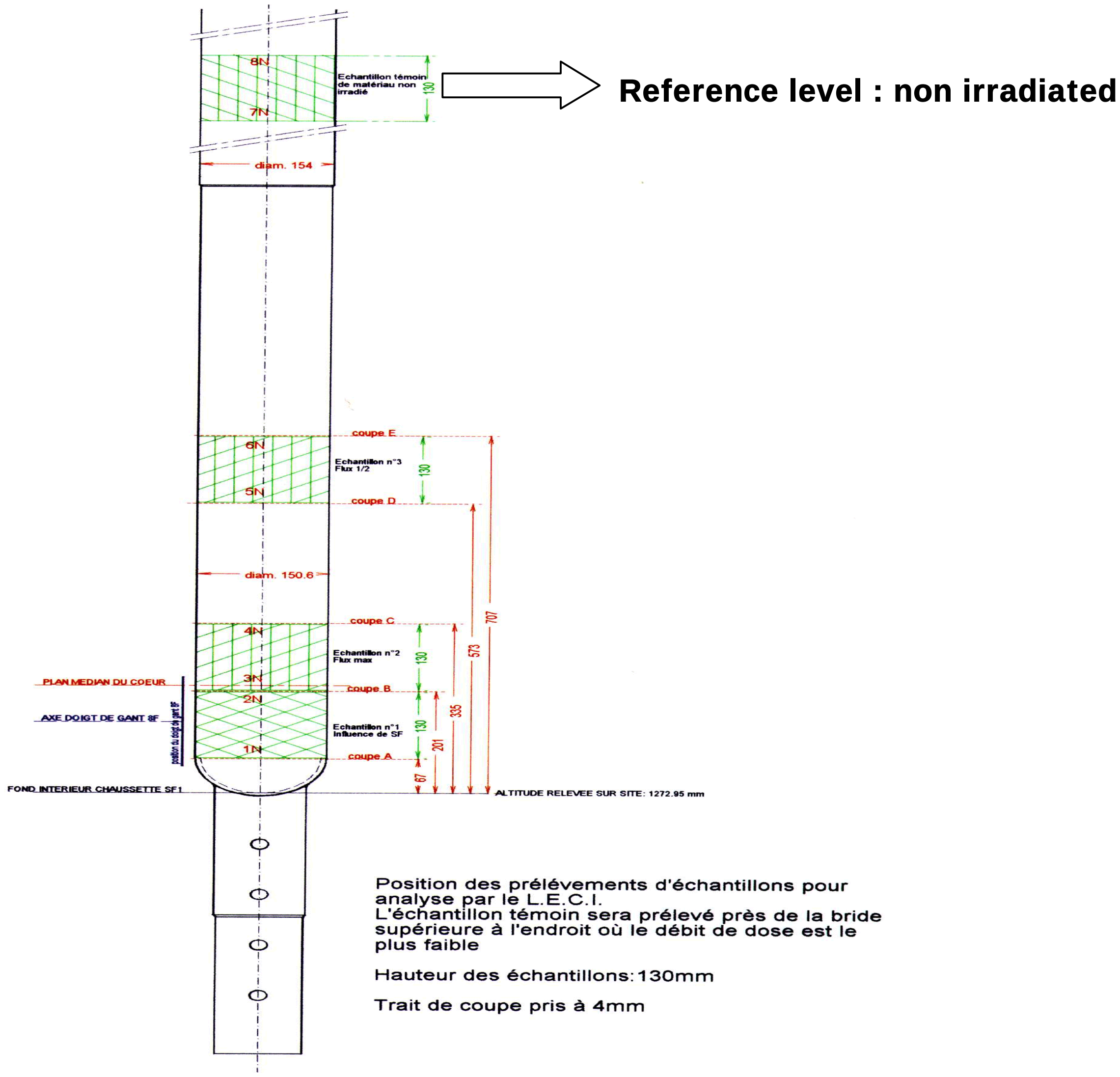


Figure 3 : Cold source shell irradiated in ORPHEE with $\Phi_{th}/\Phi_{fast} \approx 100$

The silicon content at both levels was measured by LA-OES [11]. Neutron calculations have not yet been done with the neutron spectrum of ORPHEE. However, we estimate the thermal neutron flux close to 5.8^E22 n/cm² at the maximum flux level of the cold source shell using the relation between the created silicon content and the thermal neutron fluence established by neutron calculations for the core lattice structure of OSIRIS.

The increase in mechanical strength ($R_{p0.2}$ and R_m) with the thermal neutron fluence fits well the results obtained for the core lattice structure of OSIRIS : from the non-irradiated state to the fluence 6^E22 n_{th}/cm^2, the yield strength increases by a factor 4.5 and the tensile strength increases by a factor 2.3. At the same time, the total elongation drops to a mean value of 2.5% at the fluence of 6^E22 n/cm^2.

2.3 – Influence of the spectrum index on the tensile behavior

According to the present results, the increase in strength is the same for both alloys at a given thermal fluence, but the alloy irradiated with a high ratio F_{th}/F_{fast} (≈100) has a greater loss of ductility : at a thermal fluence of 6^E22 n/cm^2, the necking at fracture Z drops to values close to 14% with

Maximum flux
F_{th} **= 5,8.10^{22} n/cm^2**

F_{th}/F_{fast} = 100, against 19% with F_{th}/F_{fast} = 1. The elongation before necking is also reduced to 1.8% with F_{th}/F_{fast} = 100 against 3% with F_{th}/F_{fast} = 1.

Previous tensile results [7] obtained on AG3-NET-O irradiated in the RHF (Grenoble) with a ratio F_{th}/F_{fast} close to 200 or 250, showed not only a drastic loss of total elongation at high fluences, but a sharp increase in tensile strength too. These preliminary results should be confirmed by precise local neutron calculations taking into account the effect of the cold source in the vicinity of the samples.

3 - CHARACTERIZATION OF COMPONENTS IN 6061-T6

As aluminum alloys of the 6000 series have not been used in the past in the French research reactors, we looked for highly irradiated components in foreign reactors. Rods in Al 6061-T6 have been identified in the BR2 reactor (SCK-CEN Belgium), stemming from a beryllium plug which has been removed in 2002 after 32 years irradiation in the beryllium matrix. The plug is cooled by light water, and the mean temperature of the rods is estimated to be 50°C. The ratio of the thermal neutron flux (<0,625 eV) to the fast neutron flux (>0,9 MeV) is close to 6.

We used 2 rods in the neutron flux and 2 rods located outside the reactor core (as a non irradiated reference) for tensile and fracture toughness tests. The mechanical characterization was performed at SCK-CEN in Belgium.

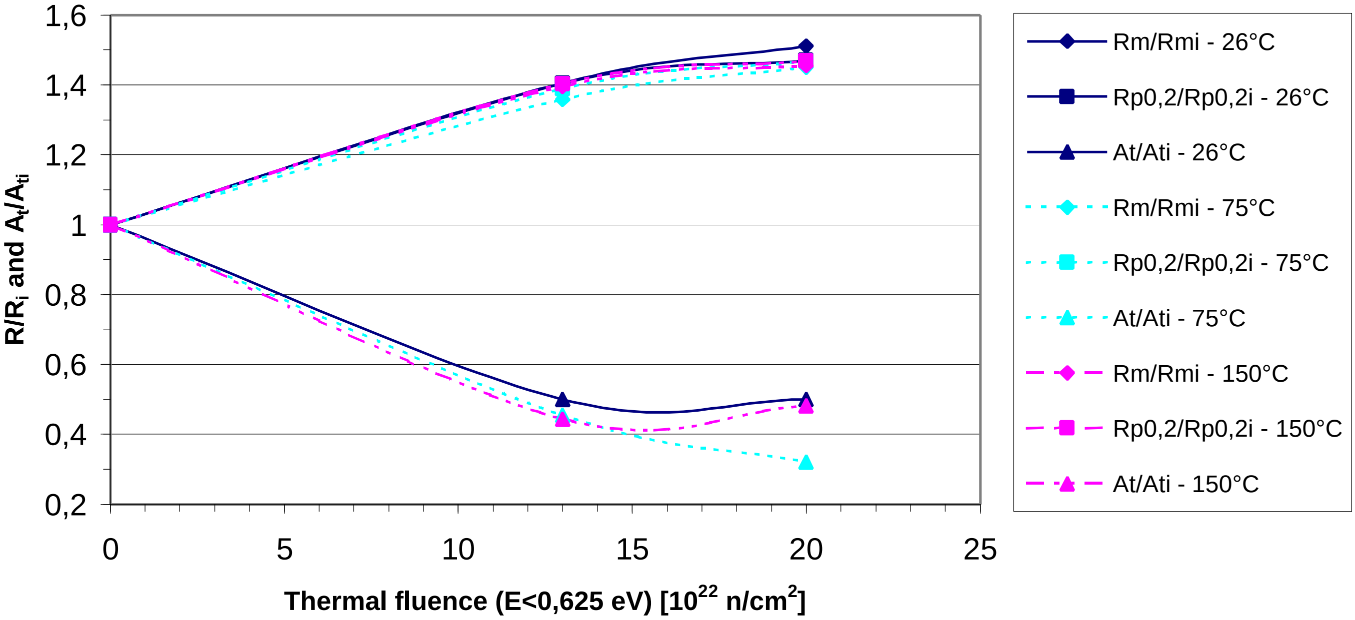
Tensile tests were performed at 2 fluences (maximum and middle value) and on the reference level ; fracture toughness was measured at the maximum fluence and on the reference level.

The silicon content was measured by LA-OES at both fluence levels of the irradiated rods and on the reference rods. Using the linear relation between the created silicon content and the thermal fluence previously established in OSIRIS, we assess the thermal fluences at both irradiated levels :

F1 = $1.3^{E}23\ n_{th}/cm^2$ at the intermediate fluence
F2 = $2.0^{E}23\ n_{th}/cm^2$ and $2.3^{E}23\ n_{th}/cm^2$ respectively for both rods at the maximum fluence

Tensile tests [12] were performed at 26, 75 and 150°C, using the standards ASTM E8 and E21, with a strain rate of $3.7^{E}\text{-}4\ s^{-1}$. The results of the tensile tests (fig.4) are in agreement with those of the bibliography : the yield and tensile strength increase with the thermal fluence by a factor which is limited to 1.5 at $2^{E}23\ n_{th}/cm^2$; and the total elongation decreases from about 9% in the non-irradiated state to values between 4 and 5% at both tested fluences. The important result is that the total elongation tends toward a limit and that the alloy keeps a certain ductility at the highest fluences. This is confirmed by measurements of the necking parameter at fracture. The ductility reaches his minimum value at 75°C.

Figure 4 : 6061-T6 - F_{th}/F_{fast} = 6
Evolution of yield, tensile strength and total elongation with fluence



The fracture toughness was measured [13] at 50, 75, 125 and 150°C, on precracked Charpy-V specimens using the standard ASTM E1820. The length of the crack was measured by the unloading compliance method. For the irradiated specimens which have a very low tearing modulus, the crack propagation is ductile unstable and does not allow for many unloadings. For this reason, the potential drop method was used together with the standard unloading compliance method.

On the non-irradiated specimens, the critical value J_{1c} of the energy integral at crack initiation increases with temperature by a factor 1.6 between 50 and 150°C. At the maximum fluence of $2.3^{E}23$ n_{th}/cm^2, J_{1c} drops to half of its initial value at 50°C, and to 1/3 of its non-irradiated value at higher temperatures ; at high fluence, J_{1c} exhibits a very small temperature dependence.

4 - OTHER IRRADIATION AND CHARACTERIZATION PROGRAMS

The mechanical properties obtained up to now are in favor of the 6061-T6 alloy, but other factors will step in the choice of the alloy for the JHR tank : welding ability, erosion resistance and sensitivity to intergranular corrosion.

The mechanical tests are being complemented by microstructure examinations : SEM examinations of the fracture surfaces, optical microscopies and EPMA aimed to evaluate the sensitivity to intergranular corrosion ; X-ray diffraction analysis of the oxide layer will be performed to determine the oxide structure in relation with the irradiation conditions of each component.

To assess the erosion of the JHR tank in the thermal-hydraulic conditions of the reactor, erosion tests are underway on the 5754-O-NET and 6061-T6 alloys with water velocities up to 20 m/s.

Welding ability tests are in progress on different grades of the 6061-T6 alloy.

We foresee to irradiate specimens in OSIRIS, in order to qualify the alloy which will be chosen for the JHR tank. A great part of these specimens will be transferred into the JHR and used for a survey program of the tank during operation.

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