Dissimilar joints for nuclear equipment & instrumentation

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1. Introduction

Nuclear fuel elements manufacture requires end closures at both ends of the cladding. In large-scale manufacture, blind end plugs are welded with specialized automated machines, the welding being performed between similar materials (e.g. resistive welding of Zircaloy plugs to Zircaloy tube, or TIG welding of Inconel plugs to Incoloy tube).

There are special situations where dissimilar joints are required, most often Zircaloy (Zy) to SS. These joints serve mostly as end plugs for experimental fuel elements (instrumented with various transducers, e.g. thermocouples, pressure sensors, etc.) or to link the expensive Zircaloy in-core irradiation section of an irradiation device to the rest of the device, made of cheaper stainless steel.

These joints have to be leak-tight and able to withstand high mechanical stress. In the case of "binary" end plugs, an axial penetration is included, to allow for passing of various instrumentation devices, e.g. sheathed thermocouple or duct for pressure measurement. It is obvious that the stainless steel remote end of the "binary" plug can be subjected to further welding / brazing operations with any kind of stainless steel parts, this being the main goal of the dissimilar joint.

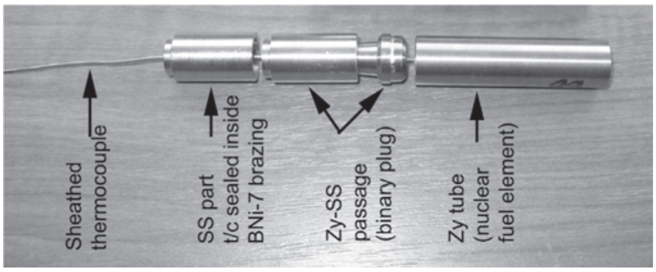

Figure 1. Instrumentation layout for a nuclear fuel element

An example of such arrangement is given in Figure 1. After completion of the binary plug as a standalone part, the rest of the joints are to be performed: Zy welding in chamber, vacuum brazing of the thermocouple in the SS part and finally TIG welding between SS facing parts.

Out of the above-mentioned Zy fuel element, Incoloy cladded fuel elements may also require instrumentation, and specialty welding is again required.

2. Devices and procedure

2.1. Vacuum induction heating device

The furnace used to obtain Zy-SS dissimilar joints is basically an induction heating equipment, composed of :

- frequency converter, a 35 kW Siemens generator, output of 10kHz, max. 600V single phase, water-cooled
- impedance adapter (single-turn secondary coil transformer and reactive power capacitor compensation).

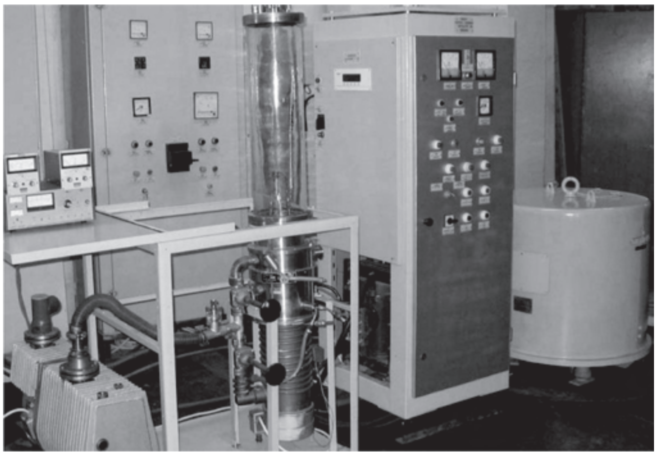

Figure 2. The vacuum induction heating equipment

- custom-made heating coil (copper pipe, usually 6 x 1 mm, water-cooled)
- control panel and automation for frequency converter, cooling and vacuum installation
- glass vacuum chamber and vacuum system (rotary pump plus diffusion pump, large range vacuum gauges)
- temperature monitors : sheathed thermocouple, electronic pyrometer, optical pyrometer with filament

2.2. Basics of Zircaloy joining to stainless steel

The autogenous brazing (without filler) of these two materials is based on the phase diagrams of the elements present in the two materials of the joint: Zircaloy and stainless steel.

Major components that have to be taken into account are Zirconium from one side, and Iron, Nickel and Chromium on the other side. With all these three major constituents, Zirconium may form eutectic, as follows [1]:

- Zr-Fe system, ~ 76% Zr (atom conc.) , liquid at 936^{o}C
- Zr-Cr system, ~ 28% Cr (atom conc.), liquid at ~ 1300^{o}C
- Zr-Ni system, ~ 24% Ni (atom conc.) , liquid at 961^{o}C

(a complete equilibrium diagram for Zr-Fe system can be easily be found in literature).

Therefore, the joining procedure would aim to keep in intimate contact the two pieces, at a temperature ~950^{o}C where the atomic diffusion is appreciable, until "critical" mixture ratio is achieved. Then a sudden raise with several tens of degrees will result in formation of a bonding liquid layer between parts (i.e. the "filler"), and its subsequent cooling completes the auto-brazing process.

2.3. Procedure

The joint is best obtained between 'father-mother' parts, carefully fit together. The 'father' part is a Zircaloy truncated cone with a 5^o taper which penetrates and fits precisely into the correspondent cone-shaped bore of the stainless steel 'mother' part. Contact surfaces roughness must not exceed 1.6 µm.

One should chose stainless steel as 'mother' part since its expansion coefficient is larger than Zircaloy expansion coefficient (17 µm/m°C vs. 6 µm/m°C at 20°C). Therefore, stainless steel part would receive better the Zircaloy cone during heating, and moreover, a fretting effect occurs at cooling down the assembly)

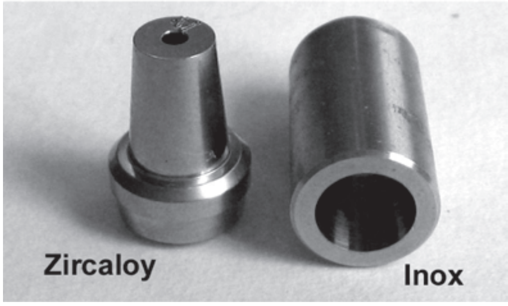

Figure 3. Parts to be joined by eutectic brazing

Clean and oxide-free surfaces are essential for this process. Therefore, several steps of preparation are required.

- ultrasonic cleaning, in a common detergent
- degrease with alcohol–acetone mixture
- pickling in acid mixture (HF 10%, HNO_3 45%, water)
- final soak in hot distilled water and drying in hot air flow

If the cleaning is correct, parts would get a bright finish and a mirror-like shine.

Pieces are put in place with a thermocouple in the axial hole, and surrounded by the heating coil (Figure 4). Care must be taken to avoid melting of the thermocouple sheath due to the contact with Zircaloy part, if the sheath is stainless steel or Inconel. A more expensive Tantalum-sheathed thermocouple surpasses this problem, and we used such a sensor. The heating coil must have a good radial symmetry, since our method does not make use of part rotation during the process.

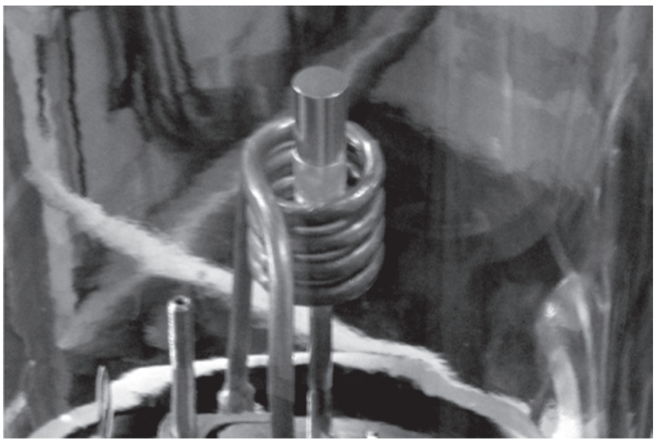

Figure 4. Close view of parts in the induction furnace

Several sample parts are provided with threads to be fixed for subsequent Helium leak test and tensile test.

The vacuum chamber is locked and the vacuum system is turned on, according to a normal procedure of a system with 2 steps (rough and diffusion pumps).

When the system is fully operational and a pressure of $5x10^{-6}$ Torr is reached, temperature is gradually increased. This is done by increasing the excitation voltage of the Siemens generator, which increases the current in the coil. Until 500^oC, vacuum quality may slightly decrease due to degassing. Hence, the temperature increase rate has to be tempered so that the vacuum level to remain in 10^{-6} decade. At 500^oC the gettering effect of Zirconium starts to be active, and one may note an increase of the vacuum quality, almost regardless the temperature increase rate. From now on, parts are gradually heated up to 950^oC.

The final heating is done setting the excitation voltage at a level determined before by experience. It has to raise the process temperature up to 1030 ± 10^oC. During this step, the operator has to keep a close eye on the 'neck' of the joint, in order to observe as fast as possible the apparition of a fine liquid 'collar', meaning that eutectic filler mixture is already formed and almost completes the joint

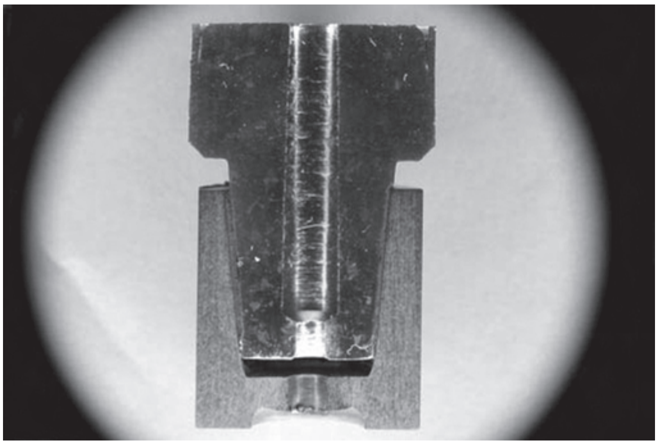

Figure 5. Macrography of Zy-SS joint. Natural illumination. Average diameter of the Zircaloy cone is 7.7 mm

According to literature and our own experience, this level of temperature has to be kept for 150 seconhds, followed by steep cooling down to 920^oC. Steep heating / cooling profile is required to mark off the 1030^oC process and therefore obtain reproducible joints. Gentle cooling is required afterwards in order to avoid solidification cracking of the eutectic zone.

3. Joint Examination and Testing

Various examinations and tests have been performed on the above-described joints, as follows:

- Macrography
- Polarized light metallography
- SEM examination
- Helium leak test
- Tensile test

Macrography was performed on cross sections of the joints, either simply polished (sand paper no. 2400), or with chemical attack specific for Zircaloy and for stainless steel, to reveal the grains. We have used a Zeiss metallurgy microscope with a Canon camera fitted on it with a remote image acquisition software.

Macrography is very useful for a general view of the whole joint, and sometimes details in macrography reveal better the cracks. It is also useful for fast measurement of the eutectic zone shape and thickness. Macrography is more relevant when the examiner has gained information and experience from the other examination techniques.

One may see bellow another macrography revealing solidification cracks in an oversized eutectic 'collar'.

Polarized light metallography is specific and recommended for Zircaloy samples. Samples were prepared through fine polishing and specific chemical attack through swabbing with acid solution (HF 10%, HNO_3 45%, water). The microscope shots have been taken with a Canon camera, at various magnifications (x 20, 50, 100, up to 200)

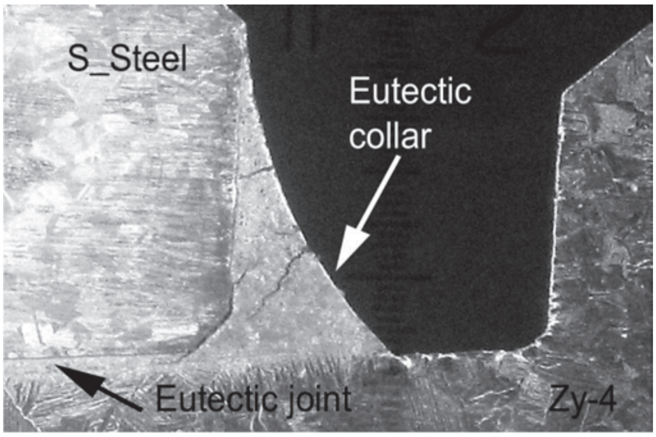

Figure 6. Macrography of a cracked area

In Figure 7, one may see three major zones: Zircaloy in the upper part, with its characteristic coloured grains (oversized by heating), then a 300 µm thick darker band, identified as being the solidified eutectic mixture, and stainless steel bellow. We may also note that the eutectic band is quite straight and uniform, at least for this magnification.

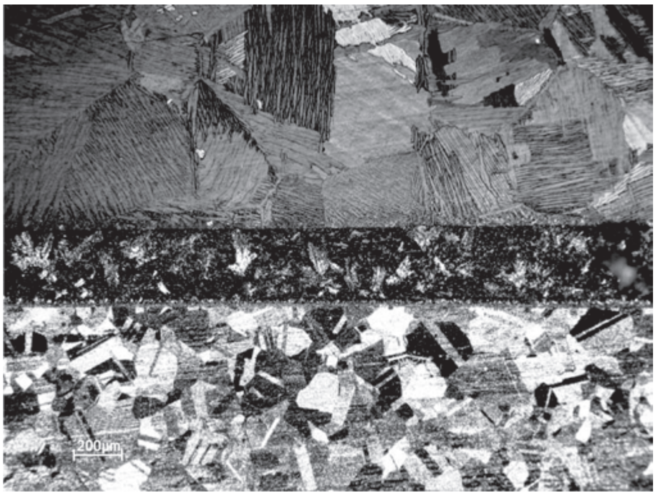

Figure 7. Polarized light metallography

Electron microscopy required finer polish (sand paper no. 4000) and no chemical attack. Two types of exams have been performed: digital imaging and elemental scanning across another sample, looking for Zr and Fe concentrations.

In SEM imaging, Zirconium atoms generate lighter areas, due to higher Z number compared to Iron that generates darker areas. In Figure 8, one may see unaltered Zircaloy in the lower-left corner, unaltered stainless steel in the upper-right corner, and a transition band with an interesting aspect.

The eutectic band is about 160-170 µm thick, and reveals solidification dendrites, emerging from SS area towards the Zircaloy area. One may also see Zirconium island among the dendrites, which denotes that in fact that dendritic solidification involves a certain degree of segregation also, towards the SS area. Anyway, detailed assessment of micro-metallurgy processes there exceeds the volume and the purpose of the present work.

The electron imaging has been completed by elemental concentration profile determination across the sample.

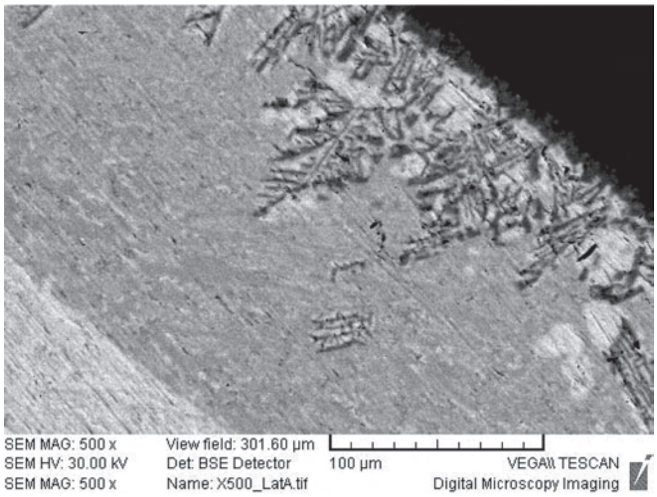

Figure 8. Electron microscopy

The concentration graphs of the two traced elements (Zr and Fe) also show the interest regions (see Figure 9) :

- to the left, the unaltered stainless steel zone, having Fe content ~ 75% (the rest up to 100% being the alloying elements as Ni and Cr in SS 304L).
- a zone of about 15 µm makes a steep transition to the eutectic composition
- the eutectic band with 160-170 µm thickness, with ~16% weight Fe and ~70% weight Zr. The rest up to 100% may be attributed to the formation and the presence the another eutectic composition, namely Zr-Ni
- Zy unaltered zone, with Zr concentration towards 95%

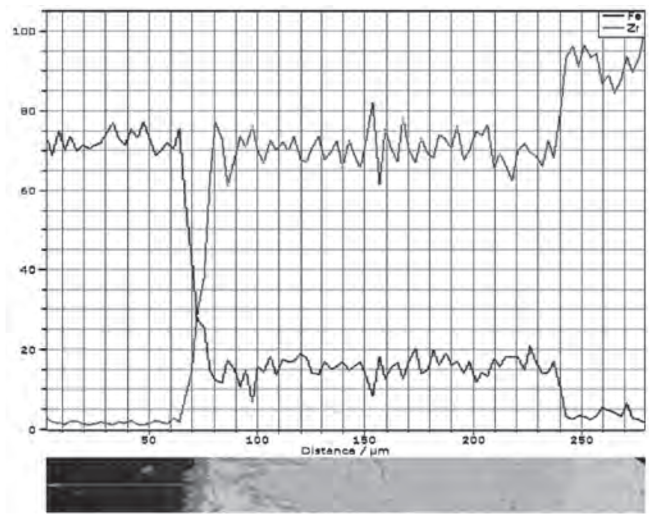

Figure 9. Elemental concentration profile (Fe and Zr)

Helium leak test is a mandatory test for this type of joint, as long as it is intended for nuclear use, and has to allow no leakage of the radioactive gases. The test showed leakage rates bellow 6 x 10^{-9} std cc / sec, which was more than satisfactory, being lower than the leakage rate imposed for large-scale CANDU fuel manufacture.

Tensile test is the last test performed in this series. It measures the global capacity of a fuel element provided with this type of plug to withstand the growing internal pressure of the fission gasses. Along with He leak test, it is a measure of the homogeneity, the strength and the soundness of the joint, in terms of regular characterization of welds.

Tensile test has been performed on an upgraded Instrom machine. Since first part of the test showed that samples were

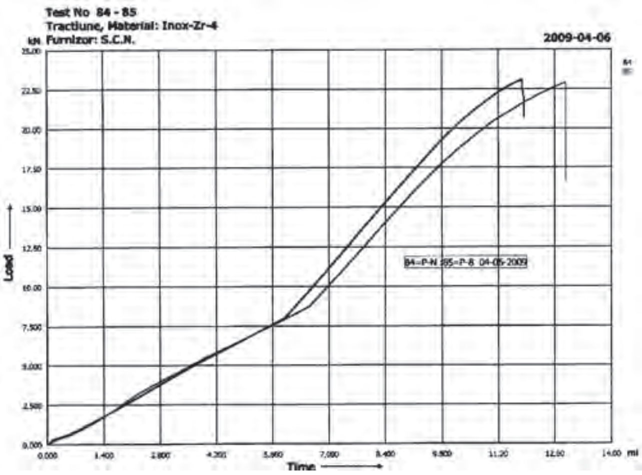

Figure 10. Tensile test diagram

not easily broken, after several minutes the load increase rate was doubled. It can be seen that the rupture occurs at ~ 23 kN for the two samples presented here.

Note that for such short samples (the length of the tested material ~ 10.6 mm) strain-stress diagram would be affected of major errors, so that we considered it useless. Therefore the “load vs. time” diagram is fully suitable for our purpose

From the aspect of the parts after rupture, some observations may be done:

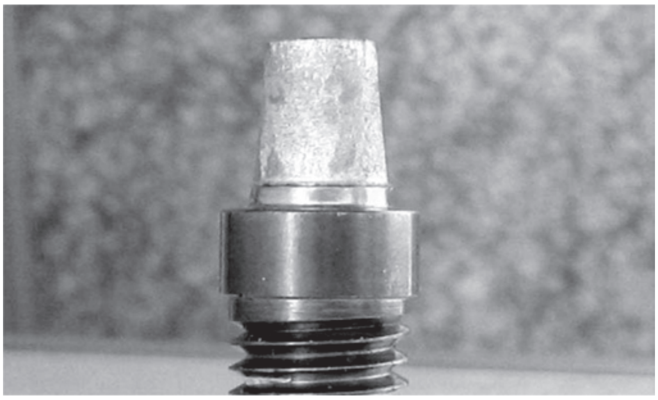

Figure 11. Zircaloy part after rupture in tensile test

After rupture, eutectic mixture remained on both parts, and there is no spot where bare base material can be seen. It means that the eutectic mixture has a very good adhesion on both parts. The rupture occurred through the eutectic zone and not along a border of the eutectic to the base material. The rupture is practically a shearing of the eutectic zone, occurred at ~7.7 daN/mm^2 (slightly lower than the values given in [1]. We also know from literature and it is no surprise that a relative thick eutectic layer to have a brittle behavior

4. Microwelds for instrumentation

Out of Zircaloy-4 another material with frequent utilization for nuclear fuel cladding is Incoloy 800, in form of thin-walled tube (~0.4 mm wall). This type of fuel element is welded with end plugs and the most trustful weld, compatible with the thermal, corrosion and nuclear conditions, is with Inconel 600 plugs. To instrument such an element, one or both end plugs need to be bored and prolonged with a Stainless steel capillary tube, 3.2 or 1.6 mm outer diameter, with internal diameter down to 0.5 mm. The capillary tube may serve as duct for a pressure measurement or passage to braze a sheathed thermocouple.

To weld capillary tubes to the Inconel end plugs may seem not difficult but we may say that it is really a challenge, since the weld has to be leak-tight down to a level acceptable in nuclear field (bellow 10^{-7} std cc/sec) and remain in such condition even after repeated exposure to high temperature and internal pressure.

We welded this type of joints both with microTIG and microPAW. We used our computer-controlled welding machine produced by Weldlogic, Ca., USA. This machine has the following main features:

- main welding module is AWS 150 based on Miller 152
- welding current DCEN (pulsed option available) up to 100 A, with 0.1A adjustment steps
- welding programs with max 9 welding levels, 60 programs may be stored in the machine memory
- rotation adjustable in 0.1 RPM steps, synchronized with the welding levels
- microplasma module, in conjunction with the main module, up to 45A, pilot arc 3-7 A, nozzles 0.6-1.8 mm

For these applications the welder has to adjust the parameters such as to get a sound weld:

- large enough to have a good dilution of the base materials (no filler used)
- with a long taper not to create stress concentration points
- small enough not to melt inside the tube opening.

One may see in Figure 12 such a microplasma weld. The tip of a ball pen is placed nearby for easy appreciation of the size.

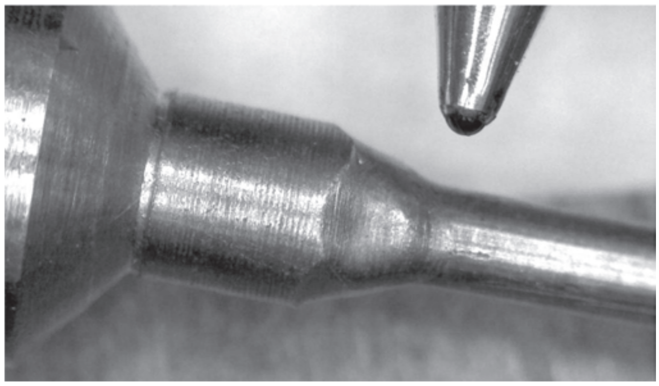

Figure 12. Dissimilar weld of Inconel cap to SS capillar

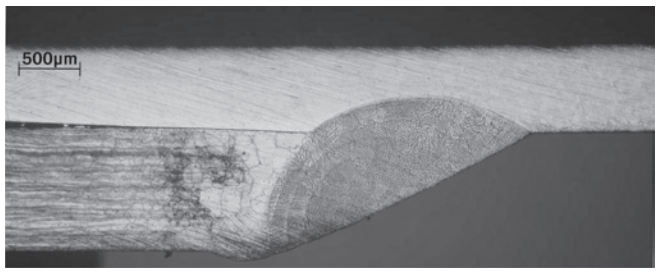

Figure 13. Metallography of a “heavy-duty” instrumentation weld.

For “heavy-duty” Incoloy capsules (heated up to 1000^{o}C simultaneously with internal pressure of 25 bars, several cycles), we TIG welded a 3.2 mm o.d. tube. One may see bellow the metallography image of this weld. We may note obvious metallurgical changes in the Inconel base metal near the weld due to the harsh treatment, but larger magnification images show no crack – the structure has preserved its integrity.

5. Conclusion

The vacuum induction device, the manufacture of parts and the working procedure itself are all fully suitable for obtaining sound joints through eutectic brazing between Zircaloy and Stainless Steel.

Several issues are essential:

- Precise 'mother-father' cones manufacture
- careful cleaning
- vacuum within the 10^{-6} Torr decade
- fine temperature monitoring and control

The obtained joints are leak-tight (Helium leak < 6 x 10^{-9} std.cc/sec) and have enough strength (rupture occurs above 2 metric tons load). Along with homogeneity, these define a "sound" joint.

Improvements must be done in temperature control, e.g. a programable system. Vacuum system has also to be refurbished with a clean turbo-molecular pump, and then the technique will be re-qualified as special manufacture process integrated in the QA system of the Institute.

This technique is a necessary completion of the TIG/PAW welding techniques already operational in the Instrumentation Lab. Together with vacuum brazing, their conjunction increases the capability of the Institute in nuclear instrumentation field, for own projects or for potential partners.

Acknowledgements

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