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Development of Electron Beam Welding Process for Welding of Zirconium Alloy to Hafnium

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ABSTRACT

Zirconium Alloys are widely used as a structural material for in-core nuclear components owing to their low neutron absorption cross-section for thermal neutrons, good corrosion resistance at high temperature, good mechanical strength and ductility. Hafnium, though exhibiting chemically similar behaviour as Zirconium, has a high neutron absorption cross-section and is a potential candidate material for control rods. Pure Hafnium is scarcely available and hence economically viable only when used in the control rod portion in

combination with other materials. Therefore, the feasibility of welding of Zirconium Alloys to Hafnium by electron beam welding was successfully studied. Zircaloy and Hafnium plates were welded in the butt weld configuration by electron beam welding. The welds were qualified by various destructive and non-destructive tests. The paper describes the process of selection of weld parameters, fixtures developed for alignment, tests conducted and results.

Keywords:— Zircaloy, Hafnium, Electron beam welding.

I. INTRODUCTION

Zirconium alloys are widely used as structural material in nuclear reactors owing to their low neutron absorption cross-section to thermal neutrons, good mechanical strength and corrosion resistance at high temperatures. Natural Zirconium contains at least 1-3% of Hafnium and nuclear grade Zirconium requires separation of Hafnium because although they have similar chemical behaviour, Zirconium and Hafnium exhibit contrast behaviour in terms of neutron absorption [1]. Hafnium thus finds useful application as control rods for nuclear reactors. The first use of Hafnium as neutron absorber dates back to the 1950s, where it was used as control rods in US submarines Nautilus & Triton [2]. Considering its desirable absorber characteristics such as good mechanical properties, resonance peaks over long range of neutron energies and sustained absorption efficiency over long periods of time [3], Hafnium is an excellent candidate for control rods. However, the costs associated with production of Hafnium have not encouraged wide usage. From an economical perspective, it is viable to use Hafnium by limiting it to the extent required by joining it with another metal which possesses good mechanical properties at high temperature and yet has low neutron absorption characteristic. Zirconium/Zirconium Alloys are suitable for this application. After performing a literature survey, it was found that the welding of Hafnium to Zircaloy is not reported. Thus, it was decided to investigate the possibility of welding Zirconium alloy to Hafnium. It is to be noted that Zirconium and its alloys are reactive and require vacuum/inert atmosphere during welding to prevent oxidation which renders the material brittle.

Electron beam welding is an established welding process for reactive metals such as Zirconium, Niobium etc. [4] as the weld is performed in vacuum and offers the advantages of weld purity, narrow HAZ and low distortion due to the low heat input. Further, the added advantage of using electron beam welding is its beam oscillation feature which aids in effective mixing of the weld pool. Due to these advantages, electron beam welding is used extensively for dissimilar metal welding [5]. This paper investigates the possibility of welding Hafnium to Zirconium alloy (henceforth, referred to as Zircaloy) in the butt joint configuration by electron beam welding.

II. MATERIALS & WELDING METHODOLOGY

2.1. Materials

The Zirconium alloy used for the study is Zircaloy-4(Zr-4). The composition of the alloy is given in Table 1. The weld samples are in the form of 4mm thick plates. Zr-4 plates are manufactured at Nuclear Fuel Complex, Hyderabad by multiple hot working and annealing cycles followed by cold rolling and annealing in the final pass. Both Zr-4 and Hafnium are machined to required dimensions so as to ensure good fitment of the weld joint and maintain squareness with respect to the width. Gap in the weld joint is very crucial in electron beam welding and a gap beyond 0.1mm is not recommended. The dimensions of both Zr-4 plates & Hafnium plates are maintained at 65mm × 50mm × 4mm.

2.2. Comparison of Physical Properties

Welding of dissimilar metals warrants the analysis of the physical properties of the metals involved. The properties that significantly affect the approach to selection of weld parameters are melting

point, co-efficient of thermal expansion, thermal conductivity. (Although metallurgical compatibility is of utmost importance, there is no literature showing the level of compatibility between Zr & Hf.). The physical properties of Zr & Hf are shown in Table 2.

Table 1: Alloying Element Composition of Zircaloy-4

Element	Composition
Tin	1.2-1.7%
Iron	0.18-0.24%
Chromium	0.07-0.13%

The weld depth of penetration in electron beam welding for any is given by equation (1)[6],

$$b = \frac{C.P}{Tk} \sqrt{\frac{K}{Vd}} \quad (1)$$

where, b= depth of penetration

T= Melting temperature

C= constant

K= Thermal Diffusivity

P= Beam Power

V= Welding Speed

d= beam diameter

k = Thermal Conductivity

Therefore, the ratio of weld penetration in Zirconium & Hafnium for the same welding speed, beam power and beam diameter is 1.4:1. Therefore, there is no vast difference in the penetration of weld for the same weld parameters. Therefore,

offset of beam is not required and the beam can be targeted on the weld joint.

Table 2: Comparison of Physical Properties of Zirconium and Hafnium

Physical Property	Zirco- nium	Hafnium
Melting point (in 0C)	1855	2233
Thermal Conductivity (in W/mK)	22.6	23
Thermal expansion co-efficient (K-1 x10-6) (at 250C)	5.7	5.9
Density (in g/cm3)	6.52	13.31
Specific Heat (J/kg K)	278	144
Crystal Structure	HCP	HCP

2.3. Determination of weld parameters

All the welds are performed on 6kW electron beam welding unit at Nuclear Fuel Complex, Hyderabad. The highest possible voltage is selected so as to achieve good penetration and narrow weld width. High welding speed is selected to minimise excess heat input. Beam oscillation is given which promotes mixing of dissimilar metals. Based on these weld parameters, bead-on-plate trials are conducted by increasing the beam current. The weld is then sectioned, polished and etched using 45% HNO₃ + 10% HF+45% H₂O and observed under an optical microscope to examine the weld penetration at different beam current values. For obtaining a homogenous weld, it is decided to weld on both side of the plates. Therefore, the current at which the weld penetration was found to be in excess of 2.5mm is selected for the sample welds. Prior to welding, both Zr-4 and Hafnium plates are degreased to remove oil contamination and then thoroughly cleaned with acetone. A

dedicated fixture is fabricated that ensures the straightness along the width of the samples. The weld is performed by clamping the plates in the fixture in a butt joint configuration and then welding along the length of the samples (i.e. along the 65mm dimensions). To ensure uniform penetration along the length and to avoid end guttering effect, two sacrificial Zr-4 blocks are placed on either side of the weld sample, wherein the upslope and down slope of the beam current are programmed to fall on the blocks and full power of the beam is incident along the entire length of the weld sample. The blocks are then removed and the edges are polished. A picture of the sample is shown in Figure 1 and the welding parameters used for the samples are shown in table 3.

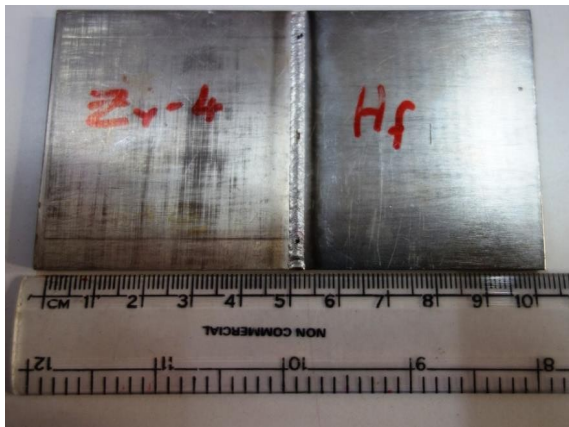


Figure 1: A Zr-4 - Hf welded sample

Table 3: Weld parameters

Voltage (kV)	Beam Current (mA)	Welding Speed (m/min)	Oscillation Magnitude	Oscillation Frequency
60	25	0.5	X=1, Y=1	75Hz

III. RESULTS & DISCUSSION

3.1. Non-Destructive Testing of weld samples

The soundness of the welds is inspected by radiographic examination. From table 2, it

is observed that the density of Hafnium is almost twice as that of Zirconium which poses a difficulty in obtaining a radiographic film with good contrast. To overcome this difficulty, one Zr-4 plate of 4mm thickness is placed directly beneath the Zr-4 plate of the weld sample. Holes are drilled on the first sample of Zr-4 to Hf weld, which serves a standard for evaluation. The radiographic film revealed that other than the drilled holes, no other indications are seen in the weld (Figure 2), indicating that the weld is sound. Hence, another two samples are made as per the weld parameters in Table 3.



Figure 2: Zr-4 - Hf weld standard for radiography with hole highlighted

The remaining two samples are also found to be of acceptable quality after radiographic test. The welds are also subjected to Liquid Penetrant Test (LPT) to check for surface defects. Both weld samples are found to be free from any indications. A picture displaying the sample after LPT is shown in figure 3.

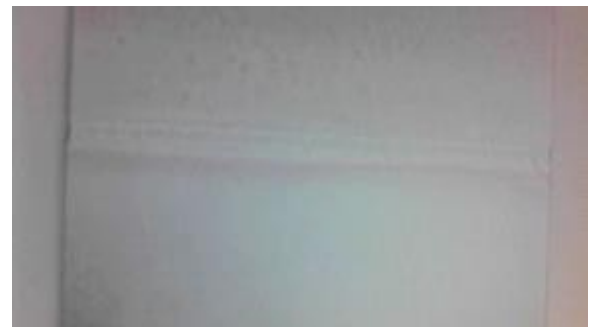


Figure 3: A Zr-4 - Hf weld sample subjected to LPT.

3.2. Weld Qualification

The qualification of weld procedure is performed as per ASME Section. IX. 2 nos. of strips of dimensions 100mm × 8mm × 4mm are cut from each of the two samples (i.e. total 4 nos.) that are sound as per radiography are then subjected to Pull-out test. It is found that all 4 tensile test samples have failed in the parent material (towards Hf side) shown in Figure 4, thereby indicating that the strength of the weld is greater than that of the parent materials.



Figure 4: Tensile Test Specimens

To check the ductility of the weld, 2 nos. of specimens i.e. one strip of dimensions 100mm × 25mm × 4mm from each of the weld samples is cut and subjected to guided bend test (Figure 5). The bend radius is 16mm (4t) and samples are bent up to 180°. Both bent specimens are visually inspected at the weld portion and later tested by LPT. No cracks or any indications are observed in the weld either in visual test or LPT.



Figure 5: Guided Bend Test Specimen

3.3. Metallographic Examination

One of the weld is cut and the weld section is polished and etched with 45% HNO₃+10%HF+45%H₂O and the area is observed under SEM. Figure 6 shows the microstructure of the weld section. There are no porosities or any other discontinuities, cracks or indications in the weld region. The weld zone appears to be sound without any defects. Micro-hardness at various locations, namely the parent material and at various locations in the weld zone are measured and indicated in Vickers Hardness (VHN). The hardness values are shown in figure 6 (with *).

3.4. Weld Purity & Corrosion

Zirconium is a reactive material which rapidly picks up oxygen and nitrogen at elevated temperatures. To test the weld purity, small specimens of 5mm x 5mm x 4mm were prepared from the weld zone and subjected to Gas Chromatography. The results indicate that the content of oxygen and nitrogen is within prescribed limits. Further, the performance of the weld under elevated temperatures and steam environment is evaluated by subjecting a sample of size 20mm x 10mm x 4mm to a pressure of 1500 psi and 4000C for 72 hrs.

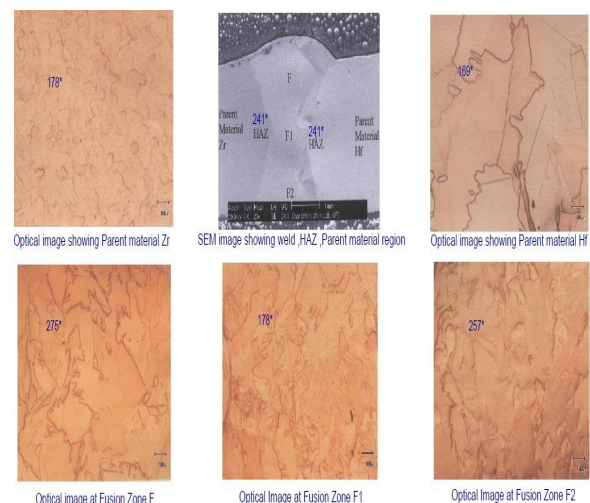


Figure 6: Micrographs of weld under a SEM

A black lustrous layer is formed on the Zr-4 plate, while a green layer is formed on the Hf plate. No white patches are observed on the sample that implies a satisfactory result. A picture of the sample subjected to corrosion test is shown in figure 7.



Figure 7: Weld sample subjected to corrosion test

IV. CONCLUSIONS

1. Hafnium is an excellent candidate as a neutron absorber with desirable properties such as good mechanical strength and can sustain its absorption efficiency over long periods of time.
2. Due to the rare availability of Hafnium, it is economical to limit the Hafnium inventory to the extent required by welding it with Zirconium alloys.
3. Electron beam weld between Zircaloy-4 and Hafnium produces sound welds with good strength and corrosion properties.
4. The scope of using Hafnium as a control rod in combination with Zircaloy structure can be explored.

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