Vacuum – The Ideal Environment for Welding of Reactive Materials

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Abstract: All reactive metals and their alloys, during welding process, are subjected to contamination by the gases present in the atmosphere which affects the strength, elasticity and corrosion resistance behavior of the weld. Although the use of shielded electrodes and inert gas surrounding during welding could reduce the difficulty of welding to a great extent, still the quality of the weld is not suitable as far as the nuclear and aerospace applications are concerned. Electron beam has been recognized as a versatile tool for welding of reactive materials as the process is carried out under high vacuum, which is being considered to be the best inert environment. A number of reactive materials have been welded by electron beam in BARC and elsewhere for aerospace, accelerator, nuclear and defense applications. A brief description of the EB welding process for some of the above materials will be given in this paper with a discussion about their post weld mechanical properties and corrosion resistance behavior.

Key words: EBW, HAZ, reactive metals, refractory metals, PHWR.

1. Introduction
Titanium and zirconium (belong to Group IV of periodic table) known as reactive materials exhibit strong affinity towards gases and react with oxygen, hydrogen, nitrogen and carbon at elevated temperature and consequently loss their ductility due to the characteristic known as the ductile-brittle transition. Contamination during the high temperature period can raise the transition temperature so that the material becomes brittle at room temperature [1][16]. Tungsten, molybdenum, niobium, tantalum are grouped as refractory materials (Group V and VI) for their extremely high melting points and higher density, have similar affinity towards gases as reactive materials [2].

For welding of the reactive metals and their alloys for the critical applications in the areas of nuclear and aerospace industries, welding atmosphere better than inert gas atmosphere is desirable. Table-I shows that Vacuum environment of the order of $10^{-3}$ mbar has proven to be contained far less gas molecules present than the purest grade of argon [3]. But no arc of conventional welding process can be sustained under this low pressure as availability of gas molecules responsible to sustain electric arc reduces. Electron beam welding process (EBW) which needs vacuum for generation and transport of electron beam is the ideal choice for low pressure welding process. In fact, invention of EBW had been taken place while trying to weld zircaloy on vacuum environment in early fifties [4]. Besides highest purity of the weld pool this high power density low heat input process has other advantages...
like high depth to width ratio, negligible heat affected zone, minimum distortion and negligible
metallurgical damages \cite{5}.

![Figure 1: 150 kV, 6kW high/ partial vacuum Electron Beam Welding Machine developed in-house at BARC in 1980.](image)

The machine has been given to a small scale industry, M/s. Siddhi Engg. Co. in the year 2004, under an MOU between BARC and the above company for the industrial exposure of this technology to common Industries.

| Table-I: Percentage of Oxygen and Nitrogen in Argon and Vacuum environment: |
|------------------------------------------|-----------|-----------|
| Welding Environment                     | % Oxygen  | % Nitrogen |
| Argon Commercial Grade                  | 0.0079    | 0.02      |
| Argon (IOLER) grade-I                   | 0.0002    | 0.00005   |
| Vacuum: 10\(^{-1}\) m bar               | 0.003     | 0.01      |
| Vacuum 10\(^{-3}\) m bar                | 0.0003    | 0.0001    |
| Vacuum 10\(^{-4}\) m bar                | 0.00003   | 0.00001   |

BARC has been actively involved in development of EB welding process for various reactive and refractory materials due to their functions in nuclear industry as well as in advanced research area like aerospace, accelerator and fusion program.

A 150kV, 6kW EB Welding machine [Figure:1] was built in house for welding process development \cite{6}. General specifications of the equipment are given below:

- Accelerating voltage: 0-150 kV
- Beam current: 0-40 mA
2. EB Welding on various Reactive and Refractory Materials:

2.1: Zirconium

Alloys of zirconium, zircaloy-2, zircaloy-4 and Zr-Nb are used extensively in the nuclear industry for their excellent properties like low neutron absorption cross section, high strength and good corrosion resistant property at elevated temperature. Furthermore, the alloys do not form radioactive isotopes and has good irradiation stability in neutron environment. They are used for the fabrication of fuel rods (Figure 3) and other components of the nuclear power reactors (Figure 2 and 5). EBW in a vacuum environment of \(<2\times10^{-4}\) mbar pressure is the best suited for this alloy as vacuum allows only a negligible amount of oxygen, nitrogen or hydrogen inclusions in weld pool and HAZ \(^7\).

2.1.1: Reflector Modules:

Zircaloy-2 (ASTM-80602) is an alloy of zirconium and tin (Sn 1.7%-2.5% w/w). A typical application of this alloy in BARC was fabrication of Reflector Modules (figure 2 & 5) for KAMINI\(^7\) nuclear reactor which consists of several hundred meters of hermetically sealed welding. These modules are made of 1.35 mm thick zircaloy-2 sheets, which were EB welded at right angle to form high precession boxes of various shape and dimensions. These boxes were used as enclosures of Beryllium oxide bricks as beryllium oxide is a good moderator of neutrons. As many as 18 nos. of these hermetically sealed modules of various shapes and sizes formed the complete reflector and moderator system for the above reactor (figure 5). These Reflector modules presently are operational in KAMINI reactor.
2.1.2: Re-entrant Cans:
These zircaloy cans were required to be fabricated for the largest nuclear research reactor in the
country for carrying out irradiation experiment on various materials[3]. These fabrication involved
seam welding of tubes and circumferential welding of tube ends to torri-spheres. After EBW process
development in BARC, the job was completed by DRDO, Hyderabad as the large chamber size of
EBW machine at DRDO was suitable to carry out this welding.

2.2: Titanium:
Titanium alloys are extremely reactive and must be thoroughly protected from gases when welded.
EBW operated in high vacuum provides an excellent welding atmosphere for all titanium alloys
whether in cast or in forged condition [8]. It can be EB welded in either in solution treated or in aged
condition and higher weld speed is preferable.
Ti-6Al-4V is a dual phase alloy contains a mixture of the two phases at room temperature is most
extensively used in aerospace industries. F-14, the famous ‘American supersonic fighter plane’ was
consist of 33 nos. of precision titanium components have been EB welded for a weld length of more
than 46 meters [9].

In India EB welding of titanium components such as propeller fan assembly, Stage-III compressor, gas
bottle adopter etc have been carried out in GTRE, Bangalore[10]. Spherical gas bottle for the SLV
(satellite launch vehicle) made out of Ti-6Al-4V alloy of different size and thicknesses have been
fabricated by KELTEC. Working pressure of the gas bottles are used to be extremely high (up to 330
bars).

3: Refractory Metals
All refractory metals are sensitive to contamination by interstitial elements carbon, nitrogen, oxygen
and hydrogen at elevated temperature which reduces their ductility. These materials, in pure form
exhibit extensive grain growth in the weld heat affected zone (Figure 6 and 7). Course grain size
materials exhibits reduce ductility that leads to cracking during welding or in fabrication. The EB
welding in high vacuum or in medium vacuum environment generally produce narrow HAZ and are
the preferred welding methods for refractory metals.
3.1: Tungsten to Molybdenum: Dissimilar Reactive Material welding:
Tungsten is the most difficult of the refractory metals to weld due to its near zero ductility at room temperature and its high melting point. A number of dispenser cathodes have been fabricated in BARC by EBW \[11\]. Dispenser cathodes made of tungsten pallet were welded to molybdenum tubes [Figure 8 and 8a].

![Figure 6: Section of Butt Joined tungsten strip (2mm thick)](image1)
![Figure 7: Section of butt joined Molybdenum sheet (2mm thick)](image2)
![Figure 8: EB welded Dispenser cathode (tungsten disk to moly tube)](image3)

3.2: Molybdenum:
Molybdenum in a fine grain condition has a ductile to brittle transition temperature below room temperature. Fusion welding results in grain growth in the heat affected zone and a coarse-grained weld fusion zone structure which results in transition temperature above room temperature \[12\]. Minimum joint restrain is necessary to avoid cracking \[13\]. It is advisable to provide root centering lip for butt joint and to allow little clearance in joint. Preheating the components near 1000ºC helps to obtain crack free weld (figure-7). EB welding of molybdenum is required to be done at lower weld speed to allow uniform wetting of grain boundaries at HAZ.

3.3: Niobium:
Niobium is being extensively used in advanced countries as resonators and cavity for beam accelerator for its excellent superconducting property. Welding trials in India are being carried out at various
research centres including BARC. The first full fledged laboratory, equipped with EB welding facility dedicated to only niobium resonator fabrication has already been set up at Nuclear Science Center, New Delhi [14]. First batch of resonators have already been fabricated. Figure-4 shows the resonator core, outer shield and cavity to be used in Linear accelerator.

Niobium when used as super conduct component in the accelerator must be welded at $<10^{-6}$ mbar oil free vacuum, as single impurity atom in the weld seam change the transition temperature of the super conductor, thus causing hotspots [15].

3.4: Tantalum Tubes for OSCOM.
Batch production of tantalum tubes have been carried out in BARC for OSCOM (Orissa Sand Complex) [3]. These tubes were to be used as ‘Protective tube of HCl generation plant’. 2mm thick tantalum sheets are rolled into cones which were welded along their seam. Every pair of these cones was finally welded circumferentially.

Weld speed for tantalum has to be set at lower side (preferably $<600$mm/min.) to avoid lack of fusion in the weld. The reason is that the metal has very high viscosity in molten stage and requires more time to fill-up the keyhole.

4. Degreasing and Cleaning Of the Components:
As all the reactive materials are carbide formers and have a marked tendency to become embrittled as a result of absorption of gas, they are required to be degreased and cleaned properly. Following chart is provided for their degreasing and chemical cleaning [1].

| S/l No | Material group | Degreasing Agent | Etching treatment (Vol %) | Pickling temp. |
|-------|----------------|-------------------|--------------------------|---------------|
| 1     | Zirconium      | Acetone or Alcohol or MEK. No Cl containing solvent to be used. | H2O-62%, HNO3-32% HF-6% | RT            |
| 2     | Titanium       | Acetone, Alcohol or MEK. | H2O-70%, HNO3-29% HF-1% | $<50^\circ$C  |
| 3     | Niobium        | Acetone, Alcohol or MEK | H2O-55%, HF-22%, H2SO4-15%, HNO3-8% | RT            |
| 4     | Molybdenum     | Acetone, Alcohol or MEK | H2SO4-5%, Cr2O3-18g/l, HNO3-4.5%, HF-0.5% | RT            |
| 5     | Tantalum       | Acetone, Alcohol or MEK | HF-0%, HNO3-10% | RT            |
| 6     | Tungsten       | Acetone, Alcohol or MEK | HF-0%, HNO3-10% | RT            |
As the degreasing & cleaning process of niobium involves hydrofluoric acid, installation of cleaning hood is recommended before starting of the production[^14].

5. Inspection and Quality Control:
Qualification testing for zircaloy (zircaloy-2, zircaloy-4 and Zr-Nb) was carried out according to ASME Boiler and Pressure vessel code section III: Nuclear Power Plant components. Although tin is extremely volatile, EPMA of EBW did not show significant loss of tin[^7].
For EBW of Niobium components, ASTM standard are followed. Niobium for the superconductor applications proper degreasing and cleaning is of utmost importance. For titanium and its alloys, applicable in aerospace industries, weld qualification are to be carried out as per Aerospace Material Qualification – AMS 2680 or Aerospace Recommended Practice ARC 1317[^10].
Welded molybdenum and Tungsten component should not be subjected with any dynamic load, as strength of welding has been noticed to be less than the parent metals. Corrosion testing and fatigue testing are mandatory for zircaloy and titanium respectively.

6. Conclusions:
Electron beam welding under high vacuum environment is an ideal tool for welding of reactive as well as refractory metals and their alloys. Many critical components made of these materials for country’s nuclear, space and accelerator programs have been fabricated using EBW process. Following points are suggested for carrying out of EB welding of the above metals and their alloy:

- All reactive or refractory metals and their alloys must be properly degreased and chemically cleaned before EB welding.
- Zircaloy for nuclear applications may be welded at<2x10^-4 mbar pressure.
- Titanium for aerospace application requires <2x10^-5 m bar pressure.
- Niobium resonators should be welded at oil free vacuum of better than 5x10^-6 m bar.
- Zircaloy-2, Zr-4, titanium, and niobium can be welded at higher weld speeds >1000mm/min.
- Tungsten, molybdenum and tantalum are to be welded at lower weld speed<600mm/min.

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