Irradiation embrittlement: Equivalent margin analysis of Linde 80 welds

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Abstract

Linde 80 was a specific type of weld flux designed by the Linde Division of Union Carbide for submerged arc welding (SAW) and used primarily in early reactor pressure vessel (RPV) designs. It was effective at protecting the weld metal from environmental degradation in the hostile environment of a RPV - high temperature (290 ℃) and pressure (15 MPa). After decades had passed, it was found that Linde 80 flux introduced a large quantity of copper (Cu) and phosphorus (P) into the weld metal. These impurities contribute to the embrittlement of the weldment by concentrating at grain boundaries when irradiated. Embrittlement makes the base metal more susceptible to abrupt crack initiation and propagation, rendering the RPV lose the cooling capability; this may eventually jeopardize the environment due to the release of nuclear products (fission products and fuel). For this reason, modern reactor designs include more advanced weld fluxes that have far less impurities. However, as many early reactors are still in operation, Linde 80 must be regularly monitored and evaluated with appropriate techniques, spanning from simple Charpy V-notch and drop weight tests to more advanced fracture mechanics analyses to ensure that it is not at risk of brittle fracture and catastrophic failure.

Background

SAW is a type of arc welding, which uses a current to generate the heat for a weld. It differs from other arc welding methods by melting a flux into a protective slag. Instead of containing the flux within a tubular electrical wire, the flux takes the form of a granular blanket atop the weld. This gives it several key advantages necessary for application in a RPV, including a higher deposition rate and improved weld consistency and quality.

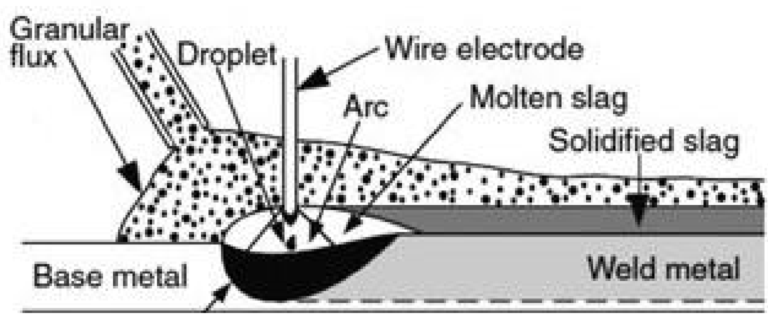

Figure 1. A diagram of the SAW process reproduced from [1].

Linde 80 welds were used in the beltline region surrounding the reactor core, nozzle to shell joints, and bottom-to-shell joints. These fit with the advantages of SAW because they are large joints that do not need high precision, yet are essential for the structural integrity of the RPV.

RPVs generally consist of A302B or A533B plates, or A508 forgings, which are quenched and tempered, low-alloy steels with primarily tempered bainitic microstructures. The composition of this base metal approximately consists of 0.05-0.2% carbon, 0.7-1.6% magnesium, 0.4-0.6% molybdenum, 0.2-1.4% nickel, 0.2-0.6% silicon, and 0.05-0.5% chromium [3]. Especially in earlier reactors, impurities were present and further introduced by the weld. The Cu concentration could get up to as high as 0.4%. The Linde 80 flux itself consists of a mixture of oxides and silicates. The non-precise manufacturing processes at the time led to the introduction of impurities such as Cu and P. The Cu impurities were particularly pronounced, because the Linde 80 weld was shielded with Cu for enhanced electrical conductivity during welding [x]. These impurities migrate to grain boundaries when exposed to neutron bombardment, which causes the embrittlement of the weld material.

Due to the problems that have been identified with the irradiation embrittlement of impurities in Linde 80 welds, they are no longer used in newer designs of reactor pressure vessels. SAW is still performed, but ultra-clean fluxes are used instead. However, due to the longevity and current age of many operating reactors, Linde 80 welds still are a present factor that must be monitored and studied to avoid degradation and failure.

The Charpy V-notch impact test is commonly used to measure the toughness of materials. This method involves creating a 10 x 10 x 15 mm3 V-shaped notch into a sample of the material, then striking it with a pendulum hammer. The energy that it takes to fracture the metal is calculated based on the height from which the pendulum falls and the mass of the hammer. The Charpy test can be used to identify the ductile to brittle transition temperature (DBTT) of the material, the temperature at which toughness sharply drops. When this test was applied to Linde 80 welds, it was found that irradiation had significantly raised the Charpy DBTT temperature.

When temperatures are high, as they are within a RPV, the energy that it takes to fracture as in the Charpy test plateaus at a higher value than when the material is at a low temperature. This is known as the upper shelf energy. For this reason, there exist strict regulations regarding the lowest possible value of the USE. In South Korea, it is stated that the maximum Charpy USE is 68 Joules [2].

Evidence of the raising of the DBTT and drop in USE in Charpy tests is well documented. One of these is documented in a 2017 evaluation of the Kori Unit 1 reactor by Lee et al [2]. This particular reactor was built in 1978 and had problems regarding irradiation embrittlement throughout its entire forty-year lifespan.

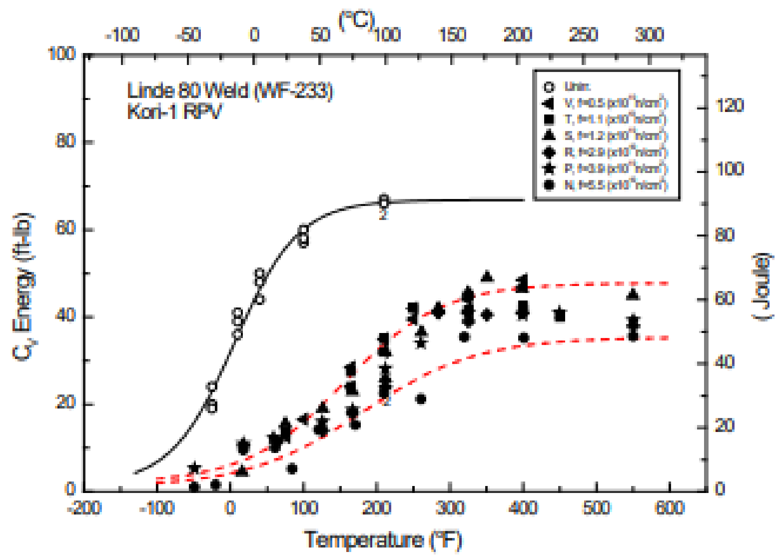

Figure 2. A graph from the 2017 paper by Lee et al. clearly showing the decreasing in energy that it took to fracture Linde 80 welds that had been irradiated [2].

Figure 2 shows that a relationship exists between irradiation and results of Charpy V-notched impact tests. All throughout the reactor, the USE had dropped well below the allowed value of 68J as per South Korean regulations. Despite this, Lee et al. concluded that there still existed a safe margin before fracture and that the reactor could have safely continued operation past its decommission date in 2017. It cannot be disputed, however, that had more precise modern methods that introduced fewer impurities been used, it would have prevented the degradation of the components and also helped reassure the public, equally important factors in reactor longevity.

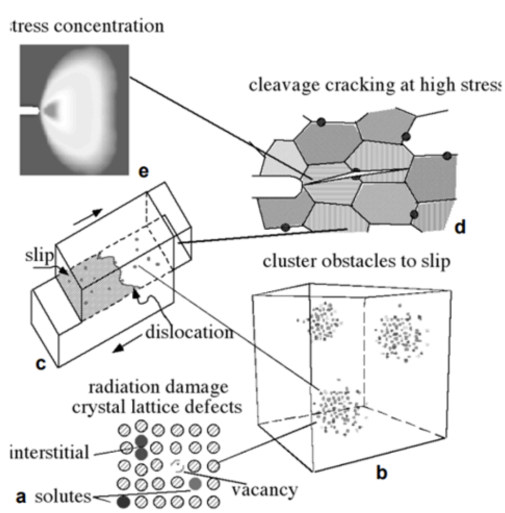

Figure 3. Graphic of the embrittlement process caused by irradiation reproduced from [3]; (a) primary radiation damage defect creation; (b) nanoscale solute and defect clusters formation (iron atoms not shown); (c) pinning of dislocations and hardening by nanofeatures; (d) hardening enhanced cleavage fracture; at a (e) stress concentration.

When Linde 80 weld flux is bombarded by neutrons as a result of irradiation, it generates lattice defects throughout the material. These predominantly consist of vacancies and self-interstitials, as shown in Figure 3a. The diffusion of these defects facilitate precipitation of nanoscale defect solute cluster complexes, solute clusters and distinct phases. This process is accelerated by irradiation (through radiation enhanced diffusion or RED), but still progresses gradually without it. In a 1986 study by Odette and Lucas [4], it was found that the maximum solubility of copper in the solid solution of a Linde 80 flux is < 0.01% at 290 degrees Celsius. Since the copper impurities introduced by the Line 80 flux well exceed that, even as higher temperatures raise the solubility limits, they precipitate out. Ordette and Lucas showed that RED accelerates this process, resulting in the rapid formation of a high concentration (≥1023 m3) of very small (~1.5–3 nm diameter) coherent (bcc) CRPs.

Instead of Linde 80 welds, current RPV designs use weld flux that tightly controls the presence of impurities like copper and phosphorus.. Examples includes Lincolnweld 888 and Magmaweld's SF401. This is done first by verifying the purity of incoming materials, then by melting to very high temperatures to burn off impurities and homogenize the chemisty. Production lines after that point are cleaned to insure no impurities get introduced. The result is a extremely clean weld flux that avoids the pitfalls present in Linde 80 welds. These designs do not require as active aging surveillance, which can make up for the higher cost in manufacturing.

Conclusion

Despite newer RPV models no longer using the weld flux, the history of Linde 80 welds can teach us about the damage that impurities can do to a materials properties. In a field such as nuclear, with the stakes as high as they are, it is paramount that the highest quality materials are used to prevent failure. In the 1970s, when Linde 80 was first developed, there was limited understand of copper and phosphorus contribution to irradiation embrittlement. As we learn increasingly more about the materials that make up a nuclear reactor system, our designs will only continue to improve.

Works Cited

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