CHARPY TOUGHNESS AND TENSILE PROPERTIES OF A NEUTRON IRRADIATED STAINLESS STEEL SUBMERGED-ARC WELD CLADDING OVERLAY*

CONF-840604--8

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DE84 014252

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ABSTRACT

The possibility of stainless steel cladding increasing the resistance of an operating nuclear reactor pressure vessel to extension of surface flaws is highly dependent upon the irradiated properties of the cladding. Therefore, weld overlay cladding irradiated at temperatures and fluences relevant to power reactor operation was examined. The cladding was applied to a pressure vessel steel plate by the submerged-arc, single-wire, oscillating electrode method. Three layers of cladding were applied to provide a cladding thickness adequate for fabrication of test specimens. The first layer was type 309, and the upper two layers were type 308 stainless steel. There was considerable dilution of the type 309 in the first layer of cladding as a result of excessive melting of the base plate. Specimens for the irradiation study were taken from near the base plate/cladding interface and also from the upper layers of cladding. Charpy V-notch and tensile specimens were irradiated at 288°C to neutron fluences of $2 \times 10^{23} \text{ n/m}^2$ ($E > 1 \text{ MeV}$).

When irradiated, both types 308 and 309 cladding showed a 5 to 40% increase in yield strength accompanied by a slight increase in ductility in the temperature range from 25 to 288°C. All cladding exhibited

*Research sponsored by the Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, under interagency agreements DOE 40-551-75 and 40-552-75 with the U.S. Department of Energy under contract DE-AC05-84OR21400 with the Martin Marietta Energy Systems, Inc.

ductile-to-brittle transition behavior during impact testing. The type 308 cladding, microstructurally typical of that in reactor pressure vessels, showed very little degradation in either upper shelf energy or transition temperature due to irradiation. Conversely, the impact properties of the specimens containing the highly diluted type 309 cladding, microstructurally similar to that produced during some off-normal welding conditions in existing reactors, experienced significant increases in transition temperature and drops of up to 50% in upper-shelf energy. The impact energies of the Charpy specimens containing the type 309 layer of cladding strongly reflected the amount of the type 309 actually comprising the specimen, falling into two distinct high- and low-energy populations with the low energy population corresponding to a higher fraction of type 309 in the specimen.

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Introduction

It has been proposed that the existence of a tough surface layer of weld-deposited stainless steel cladding on the interior of a Reactor Pressure Vessel (RPV) has the potential to keep a short surface flaw from becoming long either by impeding the initiation of extension of a static flaw and/or by arresting a running flaw. To obtain preliminary material properties typical of those needed to make such an evaluation for light water reactors (LWR's), a program has been established to obtain data on the degradation (or lack thereof) on the fracture properties of stainless steel weld overlay cladding. A recent review of the literature¹ has indicated the possibility for significant degradation of fracture properties of stainless steel weld metal under irradiation conditions relevant to LWRs. To evaluate this potential degradation, tensile, Charpy V-notch (CVN), and precracked Charpy specimens of stainless steel weld overlay cladding were irradiated to approximately 2×10^{23} n/m² ($E > 1$ MeV) at 288°C. The results of the tensile and CVN tests are reported here and compared with the properties of unirradiated cladding.

Materials

The specimens were all taken from a single laboratory weldment fabricated using the automated single-wire oscillating submerged-arc procedure for a companion program investigating structural effects of stainless steel cladding on composite 4 point bend specimens.^{2,3} The weldment consisted of a lower layer of type 309 stainless steel deposited on A533 grade B class 1 plate, followed by two upper layers of type 308 stainless steel cladding.

The welding wires for both the types 308 and 309 stainless steel were of 4 mm diameter and chosen to be representative of cladding formerly applied in industry. The cladding was deposited on plates which were 114 mm thick by 406 mm wide by 914 mm long to minimize distortion and provide an adequate heat sink. The clad plates were then given a postweld heat treatment (PWHT) at 621°C for 40 h to represent commercial practice.

The single-wire oscillating submerged-arc welding process used involved a preheat temperature of 121°C and an interpass temperature below 288°C. The welding parameters were as follows:

1. wire extension, 27.0 mm;
2. oscillation width, 19.0 mm;
3. frequency, 0.3 Hz;
4. dc amps, 500;
5. dc SP volts, 36; and
6. forward travel speed, 2.1 mm/s.

The three layers of cladding were applied to provide adequate cladding thickness (~ 20 mm) to obtain test specimens. This contrasts with typical commercial practice in which a single layer of overlay approximately 5 mm thick is applied by either multiple wire or strip cladding submerged arc procedures. Subsequent metallographic examination showed that the upper layer appeared typical of LWR stainless steel clad overlay, whereas the lower layer had incurred excessive dilution as a result of base metal melting during welding. Photomicrographs of the three passes illustrate the radically different microstructures in the finished weldment. The upper pass (Fig.1) shows a distribution of delta

ferrite in an austenite matrix quite typical of microstructures seen in good practice commercial weld overlay cladding in reactor pressure vessels.⁴ The effect of the 40 h PWHT on these materials is to partially transform the delta ferrite to sigma phase. While this is difficult to resolve in the optical micrographs, magnetic etching with ferrofluid, the use of a colloidal suspension of magnetic particles in the presence of a local magnetic field,⁵ and color staining techniques verified that the partial transformation had occurred.

The lower and middle layers of cladding, on the other hand, formed atypical microstructures due to the excessive dilution (approximately 50%) by the base metal and lower pass weldment respectively. Amounts of dilution in good practice cladding are typically in the range of 10 to 25%. The middle layer (Fig. 2) contains delta ferrite dispersed in austenite but in addition contains limited regions in which martensite is also present. The bottom layer had sufficient dilution to move it entirely from the delta ferrite-forming region of the Schaeffler diagram⁶ and into the austenite plus martensite region. Examination of its microstructure (Fig. 3) shows three distinct regions. The use of the ferrofluid magnetic etching technique and studies in the transmission electron microscope verified the lightest regions to be austenite, the light gray regions tempered martensite and the dark regions bainite decorated with $M_{23}C_6$ carbides.

Although the investigation of high-dilution cladding was not the initial aim of the cladding studies, it may well be highly germane to the question of the effects of cladding on RPV integrity. High base metal dilution of cladding, caused by inadequate control of welding procedures,

and the resulting microstructures has been documented^{7,8} in commercial RPVs. Typically, the resulting material has poorer mechanical and/or corrosion properties in the unirradiated condition; no information is available on the irradiation damage of such material. The fact that such material was included may provide insight into the behavior of substandard weld overlay cladding representative of irradiated material actually in the field.

To examine the effects of the varying microstructures two sets of tensile and CVN specimens were carefully fabricated to be contained as fully as possible within either the upper two layers (nominally type 308 specimens) or the lower layer (nominally type 309 specimens). All specimens were fabricated such that the specimen axis was parallel to the welding direction. The Charpy specimens were notched on the surface parallel to and nearest to the base metal in all cases.

Ferrite Number measurements were made on the finished Charpy specimens with a ferrite scope which locally measures the percentage of ferromagnetic material in the sample. The nominally type 308 specimens consistently had ferrite numbers of 2 to 6 (corresponding roughly to percentages of ferrite), as did the portion of nominally type 309 specimens composed of upper weld pass layers. The notched side of the nominally type 309 specimens closest to the base metal interface exhibited a wide range of ferrite numbers from 2 to greater than 30 (off scale). This wide range was presumably due to the volume of material over which the ferrite scope takes a measurement and the fact that it would, in some cases where the amount of 309 weldment was thin, include some of the type 308 upper layer clad. Optical examination of the microstructures of the type 309 layer

indicates the amounts of martensite and bainite to be between 30 to 45% and 10 to 15%, respectively.

Irradiation History

The specimens were irradiated by Materials Engineering Associates in the core of the 2-MW pool reactor (UBR) at the Nuclear Science and Technology Facility, Buffalo, New York. Two separate capsules were used, one each for the types 308 and 309 specimens, respectively. The capsules were instrumented with thermocouples and dosimeters, and were rotated 180° once during the irradiation for fluence balancing. The capsule containing the type 308 specimens reached an average fluence of $2.09 \times 10^{23} \text{ n/m}^2$ ($E > 1 \text{ MeV}$) $\pm 10\%$ during 679 h of irradiation. The capsule containing the type 309 specimens reached an average fluence of $2.02 \times 10^{23} \text{ n/m}^2$ ($E > 1 \text{ MeV}$) $\pm 5\%$ in 508 h. The fluences are for a calculated spectrum, based on iron, nickel, and cobalt dosimetry wires. Temperatures were maintained at $288 \pm 14^\circ\text{C}$ except for the initial week of irradiation. During that time, temperatures as low as 263°C were recorded for the type 308 specimens.

Results and Discussion

Tensile testing was conducted at room temperature, 149, and 288°C . Irradiation increased the yield strength of the type 309 specimens by 30 to 40%, whereas the increase of the type 308 specimens was only 5 to 25% (Fig. 4). Surprisingly, the total elongation and reduction of area of both materials increased following irradiation (Fig. 5). Tensile properties are detailed in Table 1.

The effect of irradiation on the Charpy impact properties of the type 308 weld metal representative of typical weld overlay cladding was relatively small (Fig. 6). Only a very slight upward shift in transition temperature ($\sim 15^{\circ}\text{C}$) and drop in upper shelf ($<10\%$) were observed. It should be noted for both the control and irradiated specimens that Charpy curves more typical of ferritic materials than austenitic stainless steel were observed with respect to the abrupt transition from high to low energy fracture. Fracture surfaces of selected specimens were examined in the lower-transition and upper-shelf regions. Macrographs of the irradiated type 308 specimens tested at temperatures low in the transition show flat fracture with clear definition of some of the large grains produced during welding (Fig. 7). By comparison, specimens at upper-shelf temperatures produced fracture surfaces more typical of wrought stainless steel with deep shear lips and a dull appearance. Scanning electron microscopy (SEM) of unirradiated specimens tested in the lower-transition and upper-shelf regions clearly show the transition from a cleavage or quasicleavage to a fibrous fracture mode (Fig. 8). This behavior compares well with the work of other researchers^{9,10} who have shown fully ductile fracture in as welded austenitic weld metal to occur as low as 4K, but that quasicleavage can occur in weld metal which has received a PWHT in the temperature range in which carbide precipitation and sigma formation occur. In addition to the formation of carbides and sigma phase during PWHT, the rate dependency of fracture in austenitic weld metals of decreased toughness with increased loading rate reported by Hawthorne & Watson¹¹ may also be a factor in the low temperature brittleness displayed by the impact specimens.

The interpretation of the impact results of the nominally type 309 specimens is more complicated. Since the type 309 weld pass was not thick enough to obtain specimens composed entirely of type 309 weld metal, a portion of all the specimens nominally called type 309 is indeed type 308. Macrographs of the irradiated specimen fracture surfaces show that over the range of the full Charpy curve, the portion comprised of type 309 weld metal remains bright and faceted (Fig. 9). The remainder of the fracture surface, composed of upper cladding layers of type 308 weld metal, exhibits the same behavior seen in the fully type 308 specimens.

Examining a lower-transition range unirradiated specimen in the SEM (Fig. 10) illustrates the very different fracture morphology of the type 309 weld metal just below the notch and the rest of the the fracture surface composed of type 308 weld metal. The type 309 is very flat and formed predominately by cleavage (Fig. 11) at a temperature (-32°C) at which the type 308 weld metal is still mixed mode (Fig. 12). At upper-shelf temperatures, while the type 309 and 308 weld metals can still be distinguished in the SEM, both fail in a fibrous manner (Fig. 13).

In the nominally type 309 specimens, interpreting the Charpy impact curves demands that the dual fracture properties of the type 308 and 309 portions of the material be taken into consideration. It is clear from examination of the fracture surfaces that the type 308 weld metal has a lower transition temperature than the type 309. Examining the impact data reveals a bimodal population depending upon the amount of the tougher type 308 weld metal present in the sample. The more type 308 material in the specimen, the lower the apparent transition temperature of the specimen. The compilation of the unirradiated and irradiated impact data in

Tables 2 and 3, respectively, includes the percentage of type 308 weld metal measured visually on each fracture surface. Using this percentage as a yardstick, the impact data was divided into low- and high-energy populations. The most appropriate criteria for separating the low-energy populations were arbitrarily chosen to be less than 70 and 80% type 308 weld metal for the unirradiated and irradiated data sets, respectively (Figs. 14 and 15), as these produced the most distinct difference between the data sets.

Once these populations within the type 309 data were established, the effect of irradiation was seen to be quite appreciable (Fig. 16) with both populations experiencing large drops in upper-shelf energy of up to 50% and shifts in transition temperature of up to 100°C.

The extensive toughness degradation seen in the type 309 material as compared to very little in the type 308 is probably due to the higher fraction of ferritic phases in the type 309 resulting from the excessive base metal dilution and their intrinsically higher radiation sensitivity.

Conclusions and Recommendations for Further Work

On the basis of irradiation of one weldment of stainless steel overlay under temperature and fluences similar to those at end-of-life for an LWR, it seems that very little degradation of the notch-impact toughness displayed by good quality cladding would occur. In fact, both the tensile strength and the fracture ductility were improved slightly by irradiation. It must be stressed, however, that this is only a single

case and that no conclusions, positive or negative, can be drawn regarding welding procedures or material chemistries producing material appreciably different than that studied here.

It would be very valuable to repeat this type of experiment on clad overlay produced by other methods similar to those used for existing cladding in LWR reactor pressure vessels (e.g., multiple wire or strip cladding).

Results from the highly diluted type 309 weld metal do show appreciable radiation-induced degradation of notch-impact toughness, even though both the tensile strength and the tensile fracture ductility were improved slightly by irradiation. In the few known cases where welding has produced abnormal cladding with excessive dilution in operating reactors, the radiation effects on notch-impact toughness may be cause for concern.

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Figure Captions

Fig. 1. The microstructure of the top layer of type 308 stainless steel weld overlay is typical of Reactor Pressure Vessel cladding with delta ferrite in an austenite matrix.

Fig. 2. The middle layer of clad overlay (type 308 stainless steel) includes patches of martensite (light gray) in addition to the delta ferrite in an austenite matrix.

Fig. 3. The high base metal dilution of the lowest layer of cladding, type 309 stainless steel, resulted in a three phase microstructure of austenite (lightest region), martensite (light grey) and either upper bainite decorated with additional carbides or decomposed martensite formed during PWHT.

Fig. 4. Effect of irradiation at 288°C to a fluence of 2×10^{23} n/m² ($E > 1$ MeV) on the tensile strength of the nominally types 308 and 309 stainless steel weld metal.

Fig. 5. Effect of irradiation at 288°C to a fluence of 2×10^{23} n/m² ($E > 1$ MeV) on the tensile ductility of the nominally types 308 and 309 stainless steel weld metal.

Fig. 6. Effect of irradiation on the Charpy impact energy of type 308 stainless steel cladding.

Figure Captions (Continued)

Fig. 7. Fracture surfaces of irradiated type 308 stainless steel cladding Charpy impact specimens. (a) Specimen CPC-304 tested at -60°C very low in the transition range. (b) Specimen CPC-290 tested at 150°C on the upper shelf.

Fig. 8. Scanning electron micrographs of the fracture surfaces of unirradiated type 308 stainless steel cladding Charpy impact specimens. (a) Specimen CPC-283 tested at -100°C on the lower shelf showing predominantly brittle fracture (500 $\times$). (b) Specimen CPC-298 tested at 150°C on the upper shelf showing fibrous fracture (500 $\times$).

Fig. 9. Fracture surfaces of irradiated stainless steel cladding Charpy impact specimens (nominally type 309) clearly showing the bright, faceted type 309 weld metal pass directly below the notch and the duller type 308 weld metal comprising the rest of the specimen. (a) Specimen CPL-515 tested at 0°C in the very low-transition region. (b) Specimen CPL-518 tested at 250°C on the upper shelf.

Fig. 10. Scanning electron micrograph of fracture face of unirradiated Charpy specimen CPL-516 (nominally type 309) tested at -32°C in the transition region. The dimpled area at the left is the specimen notch, the central flat portion is the type 309 weld metal, and the rough portion at the right is the first type 308 weld pass (30 $\times$).

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Fig. 6. Effect of irradiation on the Charpy impact energy of type 308 stainless steel cladding.

a

Fig. 7. Fracture surfaces of irradiated type 308 stainless steel cladding Charpy impact specimens. (a) Specimen CPC-304 tested at -60°C very low in the transition range. (b) Specimen CPC-290 tested at 150°C on the upper shelf.

Fig. 1. Continued (b)

a

b

Fig. 8. Scanning electron micrographs of the fracture surfaces of unirradiated type 308 stainless steel cladding Charpy impact specimens. (a) Specimen CPC-283 tested at -100°C on the lower shelf showing predominantly brittle fracture (500 $\times$). (b) Specimen CPC-298 tested at 150°C on the upper shelf showing fibrous fracture (500 $\times$).

a

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(b) Specimen CPL-518 tested at 250°C on the upper shelf.

Fig. 9. Continued (b)

Fig. 10. Scanning electron micrograph of fracture face of unirradiated Charpy specimen CPL-516 (nominally type 309) tested at -32°C in the transition region. The dimpled area at the left is the specimen notch, the central flat portion is the type 309 weld metal, and the rough portion at the right is the first type 308 weld pass (30 $\times$).

a

b

Fig. 11. Detailed views of the type 309 weld metal fracture surface of specimen CPL-516 (tested at -32°C) showing predominantly flat fracture with islands of fibrous tearing. (a) 100 $\times$. (b) 500 $\times$.

a

b

Fig. 12. Detailed views of the type 308 weld metal fracture surface of specimen CPL-516 (tested at -32°C) showing mixed mode fracture.

(a) 100 $\times$. (b) 500 $\times$.

a

Fig. 13. Scanning electron micrograph of the fracture surface of unirradiated nominally type 309 Charpy specimen CPL-524 tested at 177°C on the upper shelf. (a) Low-magnification view of the notch, the type 309 weld layer, and the type 308 weld layer, each comprising roughly one-third of the micrograph from left to right, respectively (30×). (b) Detail of the type 309 weld layer showing fibrous fracture (500×).

Fig. 13. Continued (b)

Fig. 14. Charpy impact energy of the unirradiated nominally type 309 cladding divided into low- and high-energy populations based on the fraction of type 308 weld metal composing the specimen ligament.

Fig. 15. Charpy impact energy of the irradiated nominally type 309 cladding divided into low- and high-energy populations based on the fraction of type 308 weld metal composing the specimen ligament.

Fig. 16. Effect of irradiation on the Charpy impact energy of high- and low-energy populations of the nominal type 309 cladding specimens.