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Protocol for Examination of the Inner Can Closure Weld Region for 3013 DE Containers

Introduction

The 2014 test plan for assessing the potential of stress corrosion cracking (SCC) of the 3013 inner can was recently issued by the Materials Identification and Surveillance (MIS) working group to determine if SCC is plausible within the 50-year design life of a 3013 storage container [1]. One of the main focus areas is a thorough evaluation of the closure weld region of inner cans opened for destructive examination (DE), which is part of the 3013 Surveillance Program. The inner can lid sections from all 3013 containers examined during DE in fiscal year (FY) 2013 and 2014 were saved so as to investigate the state of the closure weld region. An investigation of the inner can closure weld region (ICCWR) from five previous DE containers (prior to FY13) showed that pitting is a dominant corrosion mechanism in this part of the inner can [2]. Pits can act as precursors for the initiation of cracks, but the growth of cracks is dependent on numerous variables including stress levels and metallurgical conditions of the metal and surface, and the chloride concentration and humidity level at the crack tip. The ICCWR is believed to be the most vulnerable area of the inner can because of the synergistic effect of these variables [1].

The focus on the ICCWR was initiated when particulate was noted outside and within the gap of the inner lid section from the 3013 Hanford High Moisture container (HHMC) [2]. This gap, which is shown in Figure 1, results from the design of the can and is not an area examined during a standard DE. Additional analyses were required to inspect the surfaces within the gap. These analyses were performed on a few select samples taken from inner can lid sections of FY12 DE containers, which resulted in limited information about the condition of the ICCWR.

In order to conduct a thorough investigation of the ICCWR a protocol has been developed for characterizing not only the corrosion mechanism(s) but also the extent of corrosion (percentage of area and depth of attack) and the variables impacting this corrosion (chloride concentration and metallurgical condition). The protocol for the examination of the ICCWR for 3013 DE containers is presented within this report. The protocol includes sectioning of the inner can lid section, documenting the surface condition, measuring corrosion parameters, and storing of samples. This protocol may change as the investigation develops since findings may necessitate additional steps be taken. Details of the previous analyses, which formed the basis for this protocol, are also presented.

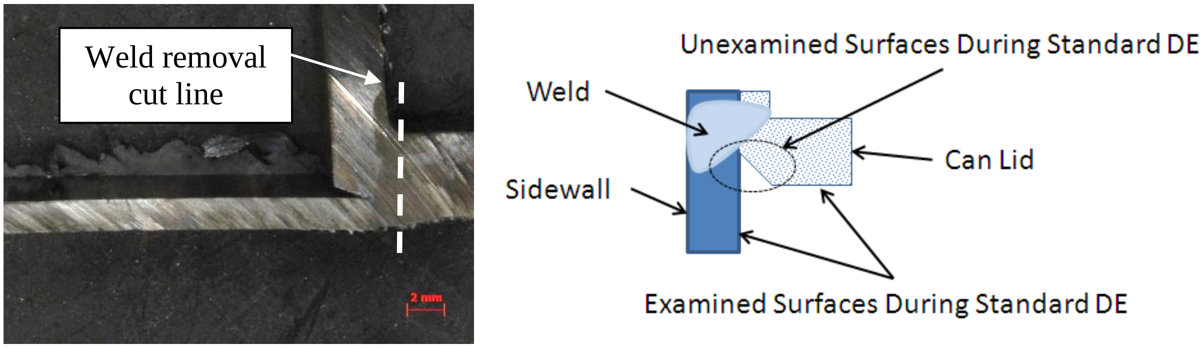

Figure 1 Cross-sectional view of a cut sample showing the gap formed between the inner can lid and sidewall and a schematic of this area showing surfaces requiring additional analysis

Analysis of ICCWR on DE Containers Prior to FY13

Although the analyses of the ICCWR prior to FY13 demonstrated that corrosion was common, the steps of the analysis were established based on the available equipment for examination of contaminated material surfaces. The equipment included a stereomicroscope (magnification up to 63x); a scanning electron microscope (SEM) with an energy dispersive spectrometer (EDS) for chemical analysis; a metallograph for examination of metallographic samples (i.e. mounted with polished surfaces) and a borescope. Location of the equipment necessitated sample transfers between gloveboxes and hoods which led to delays in work progression.

The first step consisted of cutting a small number of samples from the DE inner can lid section. A more complete sampling of the lid section was not possible due to the DE schedule and the limited capabilities. The cuts were made with the saw located in the glove box where the container examination for DE is performed [3]. Pie-shaped samples were generally cut from the inner can lid as shown in Figure 2 for the HHMC.

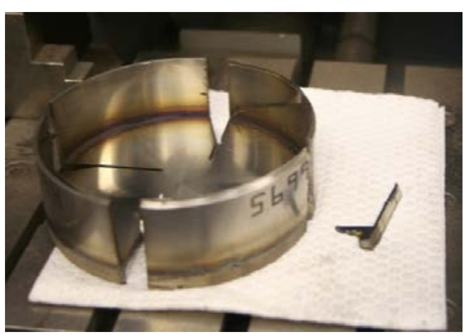

Figure 2 The inner can lid section showing location of sample cuts during the destructive evaluation of the Hanford High Moisture Can

In this first step, the samples were generally taken at orthogonal locations where the end of the closure weld as observed on the can exterior was the 0° location. The majority of the weld was from a sample so that the gap could be opened up and surface examined (see Figure 1 for the cut line). With this approach, the uniformity of the observed corrosion was unknown and the location of the most corroded area was not necessarily examined.

The next steps of the analysis were the examination of the lid and sidewall surfaces within the gap using the stereomicroscope and the SEM/EDS, followed by mounting the sample in cross section for serial examination using the metallograph [4-6]. The SEM and stereomicroscope examinations were conducted to evaluate the surface and identify regions of corrosion, especially any pitting or cracking. The EDS was used to identify the location of chlorides either on the surface or within pits. Cracking in the inner container closure weld region has not been observed to date.

Following the surface examination, serial metallography consisted of a systematic examination of the cross section of the sidewall and lid surfaces within the ICCWR as shown in Figure 3. This stepwise process, which is time consuming, provides data on the pit geometry and depth and the presence of any cracks, especially those initiated from pits. By grinding and polishing of the samples different planes were viewed. The distance between planes varies depending on numerous factors including time spent holding the sample against the grinding paper and the pressure used (distances between planes of observation are commonly 0.1 to 1 mm).

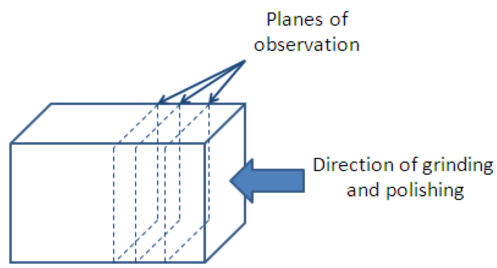

Figure 3 Schematic drawing showing the planes that are viewed during serial metallography of a sample mounted in cross section

The metallographic samples of the ICCWR were etched following an electrolytic procedure using 10% oxalic acid [7, 8]. Etching the samples allowed for the underlying microstructure to be identified. Within the ICCWR, there is the closure weld, which is an autogenous gas tungsten arc weld, the base metals of both the lid and sidewall, and the heat-affected zone (HAZ) between the weld and base metals. Each area has its own unique microstructure which will impact the observed corrosion.

A complete series of information garnered from this protocol is shown in Figure 4 for the investigation of FY12 DE6. A series of photographs showing the corrosion morphology observed on the sidewall within the ICCWR gap including a large observed pit.

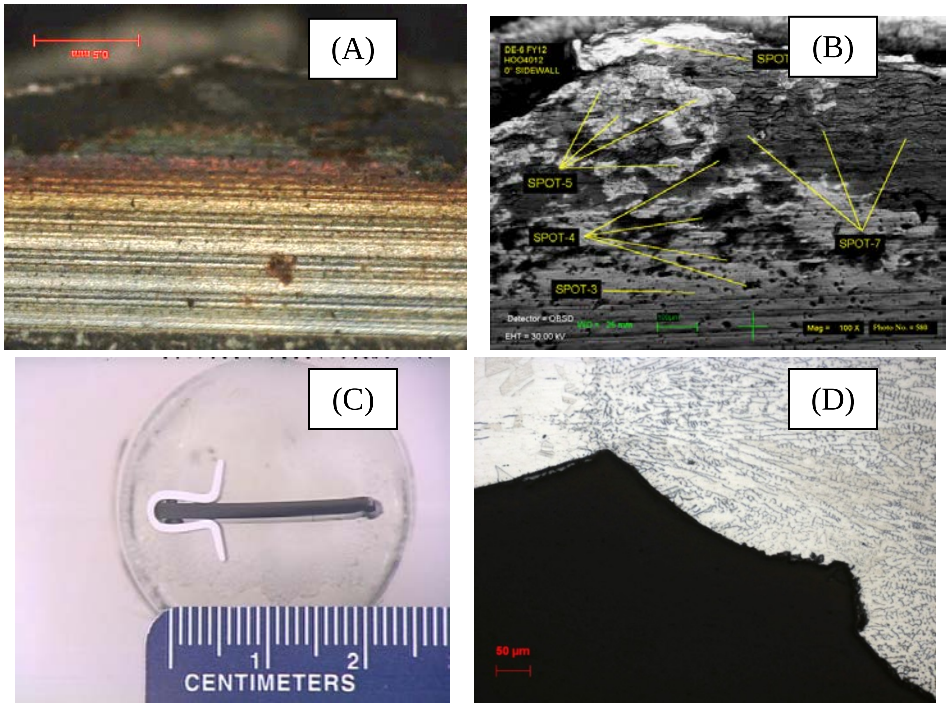

Figure 4 Pictorial view of previous protocol used during examination of FY12 DE6: (A) stereomicroscope image of sidewall; (B) SEM micrograph of sidewall; (C) cross-sectional mount of sidewall; and (D) cross-sectional view of pitting with ICCWR gap with etched microstructure

Protocol for Examination of FY13 and FY14 DE Inner Can Lid Sections

The protocol for the examination of the ICCWR from FY13 and FY 14 DE inner can lid sections will follow similar steps as those analyzed previously, but will be more developed so as to examine the entire circumference of the ICCWR and measure the deepest locations of attack. These steps include the sectioning of the inner can lid into easily handled pieces, low-magnification imaging the entire circumference of the ICCWR, surface chemical analysis of selected pieces using the SEM/EDS and wet chemistry techniques, surface depth profiling and high-magnification imaging of identified corroded areas within the ICCWR, and serial metallography of pieces with the worst corroded area for an inner can.

Sectioning of Inner Can Lid

The sectioning of the inner can lid will require two major type of cuts; quartering the can lid into four manageable pieces, and removing the closure weld just above the gap on each piece. These cuts will be made in the SRNL glovebox for DE examinations. The can lid is quartered along orthogonal lines with the 0° location at the end of the closure weld as identified on the exterior surface of the can. The lid and sidewall pieces which make up the gap will be labeled with the letter designation as shown in Figure 5. Once the lid section is quartered, these quarter sections will be stored in plastic containers with desiccant pillows to minimize the impact of moisture. The quarter sections will also be separated by plastic sleeves to minimize any surface damage. The lid and sidewall pieces from a quarter section will be store similarly whenever not in use.

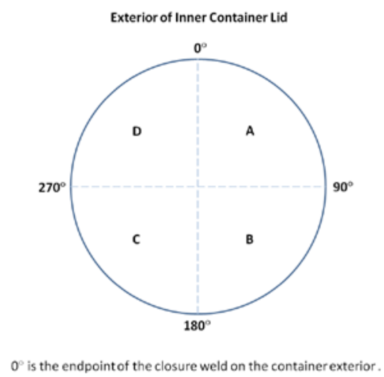

Figure 5 Labeling of inner can lid after sectioning into quarters.

The second cut to be made is to remove the closure weld so that the lid and sidewall pieces of the inner can lid quarter can be separated as was shown in Figure 1. Figure 6 further demonstrates the weld removal and the individual pieces that result from a quarter section of the inner can lid sample. Figure 6 (A) shows the individual pieces separated after a cut and Figure 6 (B) shows those pieces re-assembled with the location of weld cut line. To make the cut, the outside lid surface of quarter section is aligned parallel to the saw.

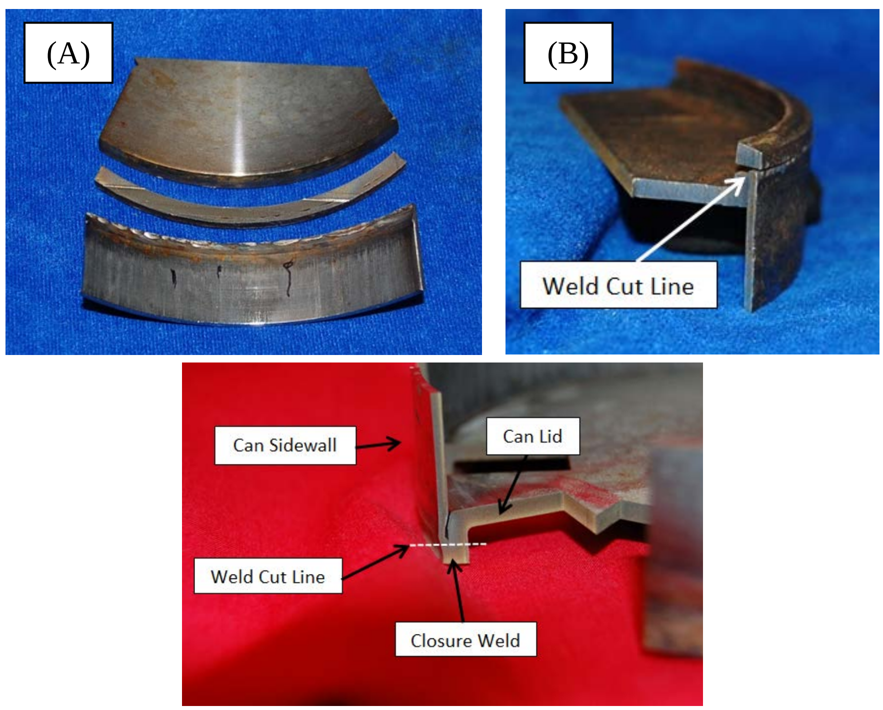

Figure 6 Removal of the inner can closure weld for separation of the can lid and sidewall quarters: (A) separated lid, sidewall, and closure weld of the inner can quarter section; (B) the pieces reconfigured into quarter section showing weld cut line; and (C) an inner can lid quarter section showing cross sectional view of gap size

The gap size of a particular DE inner can is determined by assessing the two cross-section views of each cut surface along the inner can quarter piece as shown in Figure 6 (C). The gap size is known to vary around the circumference of the can [9], which means that in some cases a portion of the gap may be removed. However, leaving too much weld metal in place will make separating the lid and sidewall pieces nearly impossible.

If the gap is small so that the top of the gap is below the outside surface of the lid (unlike the gap seen in Figure 6 (C)), the separation of the lid and sidewall pieces may require additional steps. The initial cut of the closure weld will still be made with the blade skimming along the outer side of the lid since the weld metal fragment remaining may be small. Attempts will be made to separate the two pieces. If unsuccessful, grinding away the remaining weld metal will be attempted. If the lid and sidewall pieces still cannot be successful separated, the quarter piece will be cut in half. The saw blade will make an angled cut (relative to the lid) through remaining weld metal and sidewall.

A specialized cutting jig was needed to make these precise cuts and is shown in Figure 7. The quarter piece needs to be held securely so that an even cut can be made through the weld metal. Additionally, small micrometer adjustments are needed to position the inner can quarter piece to the saw blade at a location just outside the top of the gap. The cutting jig can also be used to hold the whole can lid for quartering.

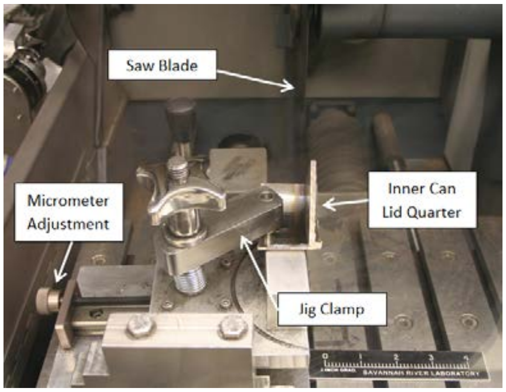

Figure 7 Specialized cutting jig for holding the inner can lid section for quartering and removal of the closure weld

Low-Magnification Images of ICCWR around Circumference of Inner Can Lid Section

A shortcoming in the previous analyses of the ICCWR was that the degree of corrosion within the entire gap was not known. The sectioning as described above opens up the ICCWR gap so that both the lid and sidewall surfaces can be examined around the entire gap. The first step in this examination is with a stereomicroscope. Low magnification (10-15x) will be used to photograph the entire circumference of both surfaces. A collage of these images, as shown in Figure 8, will assist in evaluating where the corrosion is located and the percentage of surface area affected. Figure 8 is a collage for the sidewall piece from the quarter section B from FY13 DE1. These images will be used to determine the areas for further examination with the laser confocal microscope.

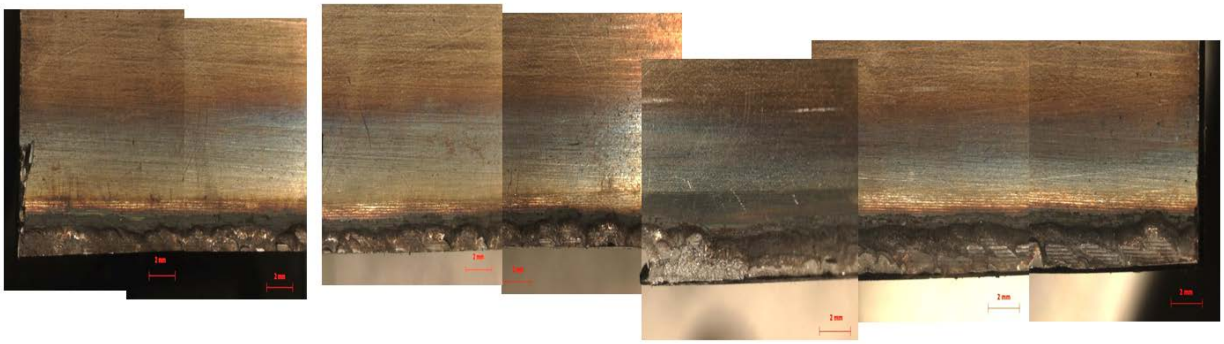

Figure 8 Collage of stereomicroscope images taken of the sidewall piece from the quarter section B of FY13 DE1 (images taken at 10x)

The collage in Figure 8 shows a missed location in the circumference as well as change in the lighting. The missed location and differences in lighting resulted from attempts at continually positioning the sidewall piece for proper alignment with the lens of the stereomicroscope. To eliminate these inconsistencies, a sample holder was fabricated so that the positioning of either the lid or sidewall surface relative to the lens are maintained constant as the surface of the piece is traversed. The sample holder has a base that connects to the stereomicroscope stage and different fixtures for holding the lid and sidewall pieces as shown in Figure 9.

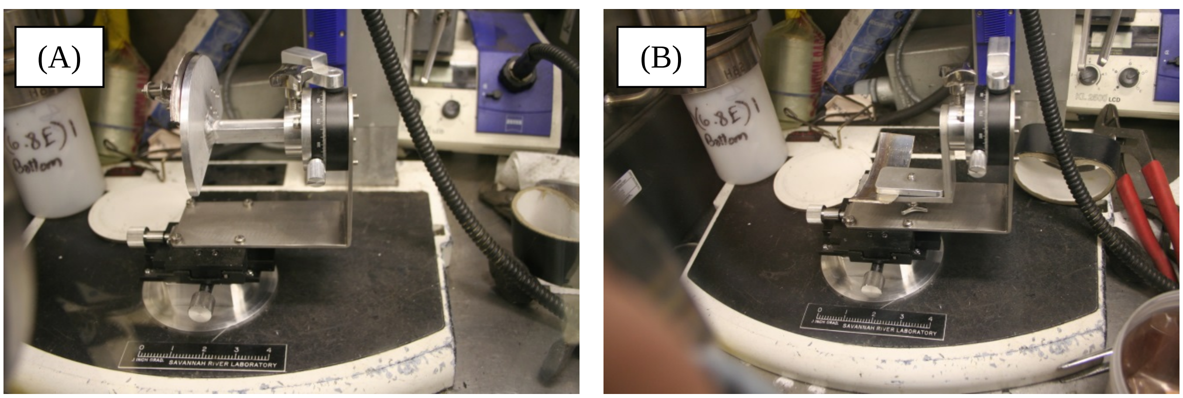

Figure 9 Sample holder for use with stereomicroscope: (A) with fixture for holding lid piece and (B) with fixture for holding sidewall piece from an inner can lid quarter section

The stereomicroscope examination is done in the SRNL glovebox (773-A, C059) where the 3013 DE examination is performed. At the conclusion of imaging with the stereomicroscope, the sections of the ICCWR will be transported to the "hot" metallurgical lab (773-A, C126) for continuation of the analysis.

Surface Chloride Determination

An important aspect in understanding a failure mechanism at the ICCWR is to know the corrosive species that contribute to this mechanism. Chlorides are typically corrosive to stainless steels and have been associated with corrosion observed on 3013 containers [10]. Both the distribution and concentration of chlorides are important in understanding the mechanism and will be evaluated. The SEM/EDS has been used to evaluate chloride distribution and form for 3013 containers where corrosion was observed, mostly on convenience cans. Both the lid and sidewall pieces from one section of the ICCWR will be sent to the SEM/EDS. The surface will be scanned to identify the location of chloride species. Once this analysis is complete, the pieces will be assessed using wet chemistry techniques to quantify the chloride concentration. The technique for solubilizing the surface chloride has not been finalized, but will consist of a soak of the surfaces within the gap of the ICCWR. This soak solution will be analyzed by ion chromatography and ion-couple emission spectroscopy to quantify the major anions and cations in the solution. Control samples will also be processed by the same technique. These liquid concentrations will be converted to an average surface concentration for the various identified species. Initially only one section will be processed for determining the concentration, which allows the other quarter sections to be analyzed using the laser confocal microscope (LCM) in the as-received conditions with weld and surface oxides still in place

Examination with Laser Confocal Microscope

The laser confocal microscope provides the capability of performing both a surface visual examination, especially for non-uniform surfaces and surface profile measurements for pit depths as well as other dimensions of surface features. Previously, visual examinations were limited to the SEM and pit depth measurements required serial metallography. Both of these analyses have significantly longer time frames for execution. The LCM will be located in contaminated metallurgical laboratory (773-A, C126).

Once a lid or sidewall piece of the ICCWR is ready for examination on the LCM, the piece will be placed into a containment box as shown in Figure 10.

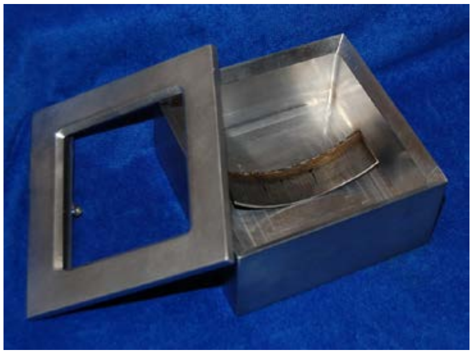

Figure 10 Containment box for ICCWR pieces with use on the laser confocal microscope

The areas of interest on the piece will be determined from the stereomicroscope examination. This box has an optically transparent window in the cover. The containment box is used to minimize the spread of contamination and to protect the microscope. Once the piece is placed into the box and positioned as needed, the box cover will be taped in place. A wax will be used to keep the piece in place and positioned. The box exterior will be decontaminated.

The examination of contaminated 3013 containers has not been performed to date with the LCM so the protocol of the examination is shown here for a quarter section taken from the inner can lid sample that was used for the boiling $MgCl_2$ testing, specifically the vapor exposure [11]. The complete progression of this analysis is shown pictorially in Figures 11 through 14. Figure 11 shows the stereomicroscope collage of the sidewall piece from this quarter section. In Figure 11 (B), the area of interest for investigation with the LCM is identified. This area was chosen since corrosion products appeared to be on the surface, the area showed a transition of surface oxides (not including the corrosion products), which are thought to be oxides resulting from the welding process, and was close to the gap opening, where previously particle accumulations have been noted [2].

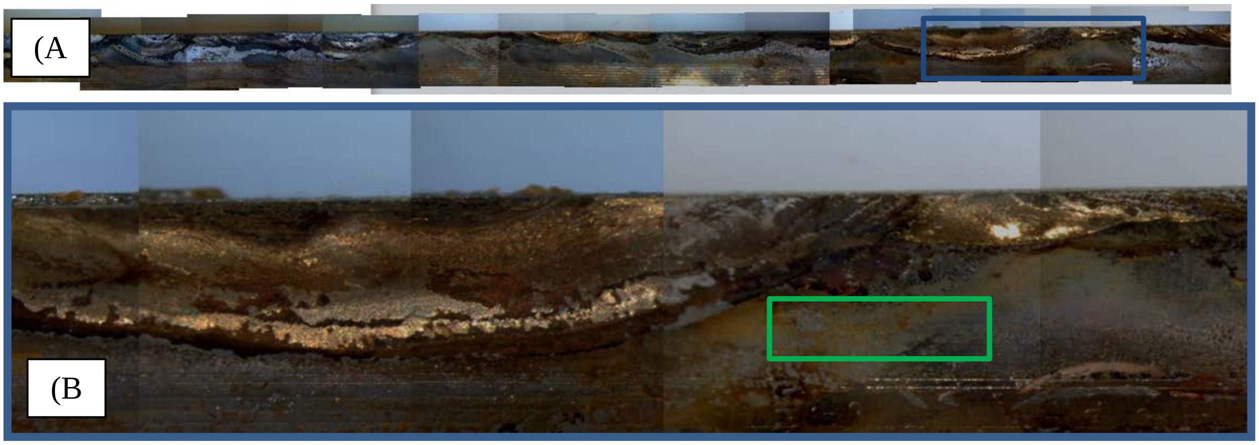

Figure 11 Stereomicroscope examination of the ICCWR from an inner can used for a boiling $MgCl_2$ test: (A) stereomicroscope collage of sidewall piece from a quarter section of the inner can; (B) expanded view of stereomicroscope collage showing area of interest (green box identifies area for LCM analysis)

For areas of interest, a series of images will be obtained to document the observed corrosion. Depending on the size of the area these will include low magnification (10x) stitched or single images. The multiple focal planes that are used in creating the images provide a completely in-focus image, which facilitates the analysis. Figure 12 shows the stitched optical image (A) and height scan (B) for the area of interest identified in Figure 11. The colors in the height scan range from a red to a blue, similar to the white light passing through a prism. The red color is the highest point and the blue color is the lowest point in the scan. The deep blue spots observed in Figure 12 (B) show pits in the surface. These pits vary both in diameter and depth (indicated by differences in blue hue). The machine marks in the surface (yellow in height scan) line above the general surface. Near the top of the image, the blueness of the whole area corresponds to the bevel that is in the sidewall of the inner can which facilitates seating of the lid during closing. The numerous pits observed in this can is a result of the exposure in the hot chloride vapors over the boiling $MgCl_2$ solution.

The next area chosen for examination is identified by the yellow rectangle in Figure 12 (A). This area was chosen since there appeared to both particulate on the surface (white particle) and pits in the surface. This area was near the oxide transition as noted by the change in coloring observed in Figure 12 (A). Figure 13 shows the single 10x optical and laser image of this area (A) and the height scan (B). Pits were then identified to observe at higher magnification (20x) and assess the pit depth (purple rectangle).

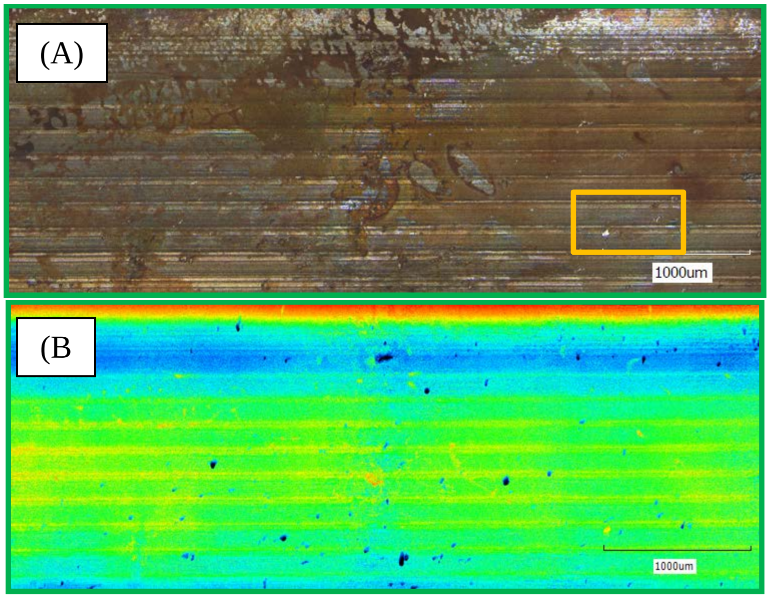

Figure 12 Stitched images from the laser confocal microscope of the ICCWR from an inner can used for a boiling $MgCl_2$ test: (A) optical image of area of interest shown in Figure 11 and (B) height scan of area of interest shown in Figure 11

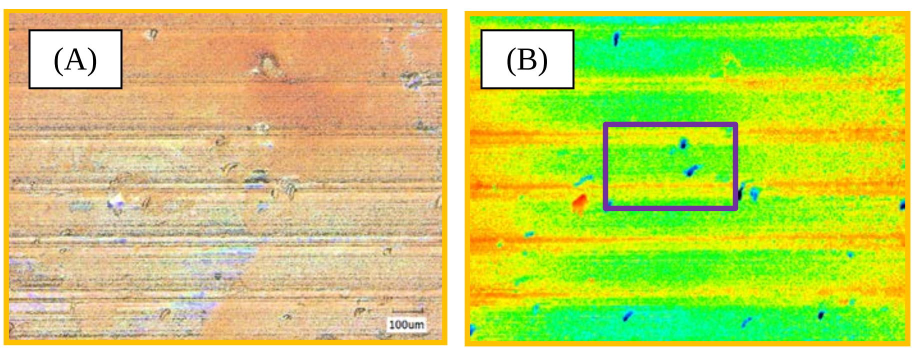

Figure 13 Single frame image (10x) from the laser confocal microscope of the ICCWR from an inner can used for a boiling $MgCl_2$ test: (A) laser and optical image of area of interest shown in Figure 12 and (B) height scan of area of interest shown in Figure 12 with purple rectangle identifying next region of interest

Figure 14 shows the results of the 20x imaging of the area of interest. As can be seen from the vertical line profile shown on the laser and optical scan (A) and height scan (B), the deepest pit in the image has a depth of approximately 22 µm. This pit measured 55 µm for the long axis and 23 µm for the short axis. Also noted from this line scan are the machine marks made in the sidewall during fabrication. These are seen as the peaks in the scan. The finer machine marks can also be seen in this line profile.

In this particular inner can, cracking was not observed; similar to the 84 examined 3013 DE containers. However, pitting was observed in the ICCWR and pits can initiate cracks beneath the surface as found during the corrosion testing in support of this plutonium surveillance program [13]. The LCM will not explicitly image these cracks if they do not break through the surface, although there could be indications in a line profile scan (i.e. Figure 14 (C)) if the crack is at the bottom of the pit and in line of travel for the laser and white light beams. To confirm if cracking is occurring serial metallography will be required.

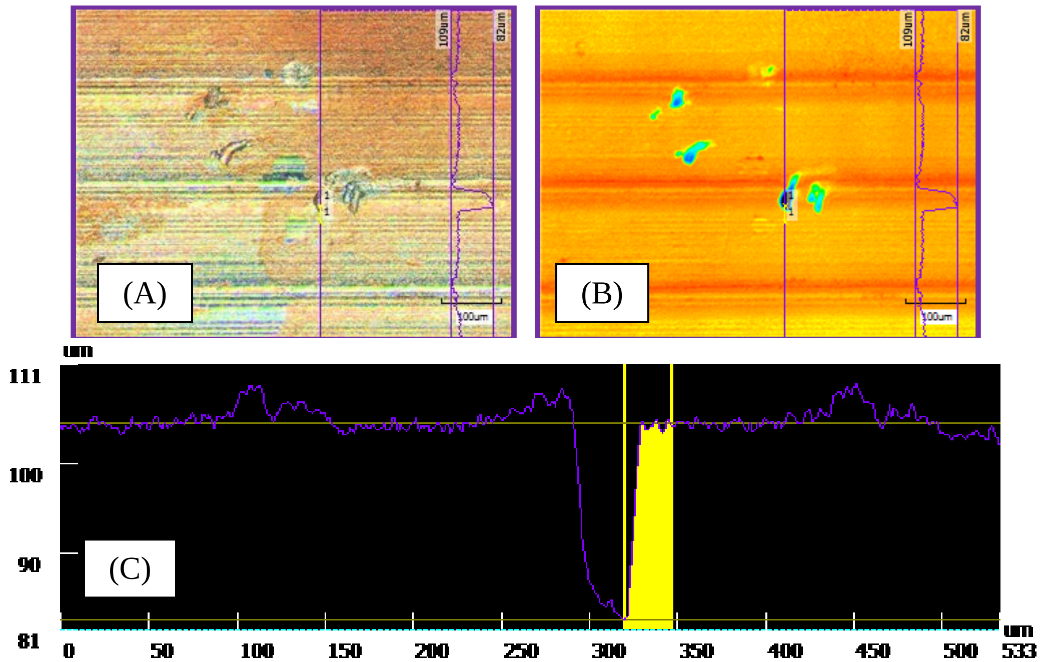

Figure 14 Single frame image (20x) from the laser confocal microscope of the ICCWR from an inner can used for a boiling $MgCl_2$ test: (A) 20x laser and optical image with vertical line profile of area of interest show in Figure 13; (B) corresponding height scan; and (C) magnified line scan showing pit depth of 22 µm

Serial Metallography of Select Pieces from the ICCWR

If cracking is not observed on the external surface, serial metallography will be used to investigate if cracks are subsurface at the most corroded area(s) as identified through the LCM. The process is similar to that described previously for the limited analysis of the ICCWR on 3013 DE containers from FY12. The number of samples to be examined will be determined based on the degree and extent of corrosion.

An inherent limitation to this examination will be to confirm that cracking has not occurred at all. If a crack does not grow through wall and remains below the surface, the only technique that would identify cracking is serial metallography. Since serial metallography is a time consuming technique, only the worst corroded area can be expected to be examined.

Conclusions

A protocol has been developed for the examination of the ICCWR from all 3013 containers examined during DE in FY13 and FY14. The MIS working group has established that this region of the inner can has many factors that increase the susceptibility to corrosion, especially SCC. The protocol was based on limited analyses of the ICCWR for the HHMC and a small number of 3013 DE containers from FY

12. A laser confocal microscope acquired by SRNL will greatly facilitate these examinations. The steps of this protocol include the sectioning of the inner can lid into easily handled pieces, low-magnification imaging the entire circumference of the ICCWR, surface chemical analysis of selected pieces using the SEM/EDS and wet chemistry techniques, surface depth profiling and high-magnification imaging of identified corroded areas within the ICCWR, and serial metallography of pieces with the worst corroded area for an inner can. The data obtained through this examination will be used to evaluate the validity of the 50-year life for a 3013 container.

Acknowledgements

The assistance and efforts of Vickie Timmerman, T. Reown and Gregg Creech are acknowledged in the development of this protocol.

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