Conf-950304--6

WSRC-MS-94-0567

Metallography of Pitted-Aluminum Clad, Depleated Uranium Fuel

by

D. Z. Nelson

Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808

J. P. Howell

Westinghouse Savannah River Company
SC USA

A document prepared for CORROSION 95-THE NACE INTERNATIONAL ANNUAL CONFERENCE AND CORROSION SHOW at Orlando from 03/26/95 - 03/31/95.

DOE Contract No. DE-AC09-89SR18035

This paper was prepared in connection with work done under the above contract number with the U. S. Department of Energy. By acceptance of this paper, the publisher and/or recipient acknowledges the U. S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper, along with the right to reproduce and to authorize others to reproduce all or part of the copyrighted paper.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831; prices available from (615) 576-8401.

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22161.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Carl: Kim's comments

"Metallography of Pitted-Aluminum Clad, Depleted Uranium Fuel" Review

The document was studied in its entirety and found to be worthy of publication in its present form. The author, however, is asked to consider the following comments for inclusion at his discretion. Items 1 & 2 should be strongly considered whereas items 3 and 4 are suggestions.

1. Figure three incorrectly maps two pairs of pits. These are the cluster of four pits located at coordinates (5.5,3).

2. Figure three depicts a scale of 2.94 cm. It is likely that this is a typo. The reference in the document to 1 inch coordinates would equate to 2.54 cm.

3. Under the section of the paper entitled 'Examination of the Mark 31 Slugs', the first paragraph refers to two unirradiated Mark 31A slugs. The discussion reads as if these slugs were chosen due to their unusually extensive corrosion. This would indicate that the absence of a radiation field correlates with higher rates of corrosion. If this is so then is an organically reducing environment possible? It may be that the radiation field has the effect of 'sanitizing' the irradiated slugs.

4. Would it be appropriate to provide a prediction of how long the slugs can be safely stored in the basin before their integrity is a concern? What will future baseline (no deionizers operating) Cs-137 rate increases be as more U core material is exposed?

11/18

METALLOGRAPHY
OF
PITTED ALUMINUM-CLAD, DEPLETED URANIUM FUEL

D.Z Nelson and J.P. Howell
Westinghouse Savannah River Company
Savannah River Technology
P.O. Box 616
Aiken, S.C. 29803

ABSTRACT

The storage of aluminum-clad fuel and target materials in the L-Disassembly Basin at the Savannah River Site for more than 5 years has resulted in extensive pitting corrosion of these materials. In many cases the pitting corrosion of the aluminum clad has penetrated in the uranium metal core, resulting in the release of plutonium, uranium, cesium-137, and other fission product activity to the basin water.

In an effort to characterize the extent of corrosion of the Mark 31A target slugs, two unirradiated slug assemblies were removed from basin storage and sent to the Savannah River Technology Center for evaluation. This paper presents the results of the metallography and photographic documentation of this evaluation. The metallography confirmed that pitting depths varied, with the deepest pit found to be about 0.12 inches (3.05 mm). Less than 2% of the aluminum cladding was found to be breached resulting in less than 5% of the uranium surface area being affected by corrosion. The overall integrity of the target slug remained intact.

Keywords: Aluminum clad, pitting corrosion, uranium metal, Savannah River Site, SRS, basin

INTRODUCTION

Aluminum clad fuel and target materials have traditionally been stored in the Savannah River Site (SRS) reactor Disassembly Basins for 12-18 months while awaiting processing in the separations facilities on site. With the suspension of processing of Spent Nuclear Fuels (SNF) at SRS several years ago to implement new regulations, policy changes, and safety upgrades, aluminum clad materials have had to be stored for extended long storage times. Mark 31A aluminum clad, depleted uranium target slugs were irradiated in the L-Reactor in 1989 and placed into the light-water filled storage basins for cooling prior to processing. Storage times for these materials have now exceeded 5 years and pitting corrosion has breached the aluminum clad, exposing the uranium core to the basin water. Corrosion of the uranium core has resulted in release of plutonium, uranium, cesium-137, and other fission product activity into the basin water environment. An aggressive Basin Management Program has been implemented at SRS to provide for control of this activity and to lead to improvements in the future storage of this Spent Nuclear Fuel. This paper discusses the corrosion of the Mark 31A slugs and presents the metallographic and non-destructive evaluations conducted on two nested slugs to determine the extent of their degradation and to better understand the corrosion process.

MARK 31A TARGET ASSEMBLY

The Mark 31A target assembly is used in the production of plutonium for defense. The assembly consists of a number of nested, short inner and outer uranium target slugs stacked inside a Uniform Sleeve Housing (USH). Figure 1 shows a cross section of a nested pair of target slugs. The depleted uranium slugs are clad with an 1100 aluminum alloy . In the manufacturing process, the uranium metal is cast, extruded, and machined into short hollow cylinders and then plated with a thin nickel layer. The plated cores are assembled into the annulus of double walled aluminum cans with integral bottoms. The aluminum cans are produced by an impact extrusion process. After cleaning, a hot die sizing process is used to metallurgically bond the can walls to the core and size the aluminum can to its final dimension. After a walnut-shell, abrasive cleaning process, the end cap is TIG welded to ensure bonding to the aluminum wall. The slugs are given a final cleaning and then an autoclaving heat treatment at 150 psig steam to provide a protective oxide coating. Finished slugs are loaded into their supporting housing tubes in the reactor area.

CORROSION OF THE MARK 31A SLUGS

Irradiated Mark 31A target slugs were moved into L-Disassembly storage in January and April of 1989. The slugs were stored in rugged stainless steel buckets in water which was periodically deionized. Water chemistry was maintained, but the primary protection against corrosion had always been to minimize storage times by sending the target materials to the separation facilities for processing. Typical storage times during normal operations have generally been 12-18 months. The extended water storage of the Mark 31A slugs caused by the suspension of processing by the separations facilities has resulted in continued wet storage, now exceeding 5 years. The Disassembly basins were not originally designed for continuous operation of the portable deionization system to provide the high quality necessary for the long term storage of aluminum clad materials. During the first year of water storage, there was no significant increase in basin cesium-137 activity. About January 1990, the cesium-137 activity in the basin water measured about 35 dpm/ml. By January 1991, This activity had risen to 77 dpm/ml. This increase in activity was an indication that the 30-mil aluminum cladding was beginning to be penetrated. The penetration of cladding by pitting corrosion of the aluminum exposes the uranium metal core and allows cesium and other fission products to dissolve in the basin water. About June 1991, the activity began to show a much higher rate of increase toward the Administrative Limit of 500 dpm/ml. By operating the deionizer in the basin in a more continuous fashion and by using a special zeolite deionizer built specifically for cesium control, the cesium levels have been maintained in the 400 dpm/ml range, well within the Administrative Control Limit. When the deionizers are not operating, the cesium-137 activity increases in the basin at a measured rate of about 2 dpm/ml/day.

By mid-1991, significant corrosion product was visible from a majority of the slugs in the stainless steel buckets. Areas of white hydrated aluminum oxide from corrosion of the aluminum cladding and other areas of black and yellow uranium oxides were clearly visible on the slugs. Figure 2 is typical of corrosion on the slugs in the stainless steel buckets in L-Disassembly Basin. This and other evidence of pitting corrosion on aluminum components in the basins led to the initiation of basin and laboratory corrosion testing in late 1991 to better understand the corrosion in the basins. Details of the electrochemical and component immersion tests conducted on the aluminum alloys used for fuel and target materials stored in the basins at SRS have been previously reported.[1,2]

Results of early coupon tests in L-Basin and component immersion tests in K-Basin indicated that the 8001 aluminum alloy clad used on high temperature fuel tubes could be breached in about 45 days and the 1100 aluminum alloy clad, like that of the Mark 31A slugs, penetrated in six months. These early tests were not conducted with stainless steel galvanic couples, but observations of these couples acting on the vertical hanging fuel assemblies and in laboratory tests indicate that the stainless steel/aluminum alloy couple accelerates the pitting corrosion process in basin water. In actual basin storage operations, the aluminum slugs are in direct contact with the 304L stainless steel bucket and the buckets are stacked on a large stainless steel plate on the bottom of the basin. The buckets have recently been individually overpacked in another vented stainless steel box designed to contain the corrosion product. The boxes have been insulated from the steel baseplate by a polymeric sheet material place between the overpack box and the stainless steel baseplate. Since pitting is an electrochemical process, the high conductivity of the basin water compared to totally deionized water would enable higher current flows and result in higher corrosion rates. The conductivity of the K-Basin during the early time of testing was about 180 μS/cm with impurities like chlorides in the 6-9 ppm range. These conditions compare to about 1 μS/cm and 20 ppb chlorides for the Receiving Basin for Off-site Fuels (RBOF) at SRS where aluminum clad materials have been stored for times exceeding 11 years without visible corrosion.

An excellent description of classical corrosion of uranium clad is provided by the work done in the late 1950's.[3] For aluminum clad fuel, once the clad has been penetrated by pitting corrosion, the compounds formed by the nickel bonding layer may become slightly anodic to the aluminum and result in undercutting of the cladding by galvanic corrosion when exposed to the basin water. When uranium is metallurgically bonded to aluminum by an intermediate material like nickel, it corrodes somewhat more rapidly than bare uranium, but the increase in rate is more than offset by the restricted area of attack. The corrosion of uranium in water can be expressed by the equation

$$U + 2H_2O = UO_2 + 2H_2 \qquad (1)$$

The corrosion can be divided into two stages: an initiation stage, corresponding to the induction period observed in the corrosion of bare aluminum, and a propagation stage. The initiation stage usually is an unpredictable length. There is generally absence of any significant swelling, but occasional bubbles of hydrogen may be evolved, and the pinhole in the aluminum may be discolored by small particles of UO_2. The propagation stage is characterized by the growth of a blister at the pinhole. Once swelling starts, the blister grows at a fairly steady rate until the accumulated uranium oxide causes the cladding to split. After the cladding splits, the UO_2 is released into the water and a larger area of the metal is exposed to attack. If the uranium core is of sound metal and the bond layer has no flaws or discontinuities, the blister is usually localized at the pinhole and has a mountain-like profile.

If the uranium contains stringers of voids or rolling seams, diffusion paths are provided for the hydrogen resulting from attack at the pinhole. Because of the small diameter of such flaws in uranium, the hydrogen can diffuse through them more rapidly than water, steam, or air. When the hydrogen encounters a site susceptible to attack (not protected by oxide), uranium hydride can be formed. This hydride attack is characterized by the appearance of a blister at a distance from the original pinhole. Since uranium hydride forms rapidly and has a lower density, the hydride blister almost always grows more rapidly than the original blister at the pinhole in the cladding, and the cladding usually splits first at the hydride blister. The splitting of the hydride blister exposes uranium hydride to water, with the formation of UO_2 and hydrogen, and simultaneously exposes a large area of uranium to attack. The hydride attack is generally more rapid than the direct attack by water.

EXAMINATION OF THE MARK 31 SLUGS

Two Mark 31A Slugs stored in the L-Disassembly Basin for over 5 years were identified from the bi-monthly basin surveillance photographs as having corrosion which indicated a breach of the aluminum clad. Each of these slugs was stored in a separate stainless steel slug bucket and was covered by voluminous amounts of white aluminum oxide and black and yellow uranium oxide corrosion products. Much of the loose product fell from the slugs when the slug buckets were earlier moved into overpack boxes, but the black oxide was still very visible around the pits in the aluminum clad. The slugs were removed from the buckets beneath the water using special tools, brought to the surface where they were cleaned using atomic wipes, and shipped to SRTC. These slugs were un-irradiated, but were stored in the same general area with the buckets of irradiated slugs. One of the two slugs from the basin sent to SRTC was removed from the top row of slugs seen in Figure 2. There was some contamination associated with the slugs from storage in water which contained cesium and other fission products. A radiation reading of 3-5 mR was detected on the specimens out of the water.

Once in the metallography lab the specimens were photographed in the as received condition using a 35 mm camera with color film. After this documentation, the slugs were decontaminated using a 50% solution of phosphoric acid and water. This cleaning removed the contamination and permitted the slugs to be handled like clean samples. The slugs were marked into a grid of one inch increments in the circumferential and longitudinal directions to make pit identification easier and then re-photographed. The number, the size, and the locations of the pits on each slug were mapped and documented.

Prior to the metallographic examination of the slugs, a conventional nondestructive technique, gammography, was used to evaluate the condition of the uranium billets. Single-wall, conventional gammagraphs were made on each element . Tangential gammagraphs were made to evaluate the nature and extent of the corrosion and blistering of the aluminum cladding. The conventional and tangential gammagraphs were used to evaluate the proper sites for rough sectioning and final specimen locations for the metallographic evaluation.

Metallographic Preparation

Due to the many corrosion sites and the size of the Mk-31A billet, a preliminary sectioning was performed. The billet was sectioned into four ring sections approximately two inches in length to allow precision cutting in localized areas, This sectioning was performed using a power hacksaw with a blade of 12 teeth per inch. Specimen cooling was added during the cutting to eliminate heat build-up as well as to eliminate gumming of the blade.

Each ring section was then evaluated for the best possible sites to obtain representative corrosion specimens. The sites selected were areas that could be completely enclosed in a 1 1/4 inch (3.17 cm) mount or could be halved into two mounts. Areas were also selected in the transverse and the longitudinal orientation. Fifteen specimens were cut from the four ring sections using an aluminum oxide abrasive blade on a saw running at 6000 rpm. Water was used as a coolant during the cutting process. Any deformation that occurred from the sectioning was removed in the rough grinding process.

The specimens were then mounted using a hot mount phenolic resin. To allow the possible flow of resin into voids and aluminum or uranium oxide regions, a two minute preheat step was used. The oxide areas were not initially filled with a suitable cold resin because it was not known what effect filling these regions with resin would have on the evaluation of the specimen.

Rough grinding of the specimens was performed using silicon carbide grinding paper and water. Progression through finer grits of paper was performed. To remove any possible deformation from the sectioning, an initial grinding was performed using 220 grit paper for 90 seconds with a specimen force of 25N (5.6 lbs.). After the rough grinding, plane grinding, using silicon carbide papers with a grit of 320, 500, 1000, 1200, and 2400, was used. Each plane grinding step was performed with water for a 90 second interval. Specimen force began at 25N (5.6 lbs.) for the first 70 seconds. The load decreased to 20N (4.5 lbs.) for the last 20 seconds of the grinding process to decrease the possibility of deformation and to ensure planeness.

When plane grinding of the specimens was complete, diamond polishing of the specimen was performed. Diamond suspensions were used with a medium nap woven polishing cloth. Initial polishing was performed using a 6 micron diamond suspension on the wheel at a speed of 150 rpm. Specimen force in this step was critical. Too much load caused diamond particle imbedding and too little would not remove plane grinding scratches. A load of 16 N (3.6 lbs.) was found to be the optimum force on the specimen pressure. Progressive polishing steps were performed using 3 micron diamond suspension followed by a 1 micron diamond suspension. An alcohol based lubricant was used in all of the polishing steps. All grinding and polishing residue was collected and treated as contaminated waste material.

RESULTS

Metallographic Preparation Techniques

In performing metallographic analysis for the first time on new materials such as the case on these aluminum clad, uranium core Mark 31A slugs, there were a number of problems encountered and techniques that had to be developed during the process. These are presented in this paper to make future investigations by others aware of the difficulties and enable one to benefit from the solutions that were developed.

The critical factor in the specimen preparation was found to be the forceapplied to the specimen against the polishing and grinding wheels. Preparation effects were a constant problem. Smearing and diamond particle imbedding was evident when specimen forces were too high. When specimen loads were too light, plane grinding scratches were not completely removed. A few plane grinding scratches left on the specimen were deemed better than allowing the smearing or imbedding that would occur from over pressure on the specimen.

Another major problem that occurred during the specimen preparation was caused by the extreme brittleness of the uranium oxide formed in the corrosion sites. Whenever uranium corrosion occurs, large amounts of oxide corrosion product form. Oxide in these specimens was extremely brittle and fragile and would fall out often, contaminating the polishing cloths. "Wild scratches" (scratches caused by these oxides) were observed in the final polishing steps due to this oxide fall-out and contamination. Initially, all specimens were prepared without any special stabilizing resin added to the oxide regions. At a later time, some specimens were prepared a second time after a cold resin was vacuum impregnated in the oxide regions. This process stabilized the oxide regions, but caused these regions to be obscured from view. It was found that the addition of the cold resin process was not suitable in the application.

An attempt was made in a final polishing step to use an oxide polishing solution on a medium nap chemical polishing cloth. This process performed well for the aluminum cladding, but caused the uranium core to oxidize and discolor during the process. This obscured the surface. This would have increased the quality of the specimen surface for microscopy but the discoloration prevented this step from being successful.

Another major problem encountered in the preparation of these specimens was the heavy oxidation of the uranium core after the clean surface had been prepared. The oxidation layer did not form evenly across the surface so a splotchy appearance developed. Due to this fast oxidation, as soon as the particular specimen preparation was completed, the specimens were stored in an ethyl alcohol bath until ready for microscopy. This did not completely stop the discoloration of the surface but slowed its rate long enough for photography to be performed.

With the overall goal of this metallography being to understand more about the corrosion process and the extent of the oxide buildup beneath the clad and between the clad and the core, the specimen surface quality was sometimes sacrificed when the particular area of interest was the best for viewing and photography. If material characterization is the ultimate goal, it is recommended that the sample be initially mounted in phenolic hot resin and then impregnated with a cold resin in the voids to stabilize the oxide regions of the specimen.

Mapping of the Corrosion Sites

The nondestructive examination of the slugs using iridium 192 gammagraphs confirmed that both the aluminum cladding and the uranium metal were degraded. It also revealed some areas of metal loss extending beneath the surface of the aluminum clad The high density of the uranium core made this examination extremely difficult. Thirty minute exposure times were necessary and even this exposure did not differentiate between the thin aluminum clad and the uranium. This technique did not provide significant additional information beyond the visual information acquired by using conventional photography.

The Mark 31A nested assembly shown by the arrow in Figure 2 was sent to SRTC where it was de-nested. The inner target slug was free of pitting corrosion. The outer target slug is shown in Figure 3. Many of the pits are clearly visible on the outer surface of this target slug. The dark areas represent pits which have penetrated through the 30-mil aluminum cladding and into the aluminum core. Note that most of the pits appeared to initiate in scratches on the outer surface caused by the removal of the slugs from the ribbed outer housing. Scratches in the protective oxide coating on the aluminum make pit initiation easier. An inspection of the scratched areas found a number of small white aluminum oxide nodules located within the scratches. These sites are the initiation of most pits. Once the pits initiate, they continue to grow larger until they coalesce, in many cases, as shown on this photograph of the slug and form large breached areas of the clad.

The one inch grid marked on the slugs was used to provide accurate measurements of location and size of the pits covering the surface of the outer slug. The measurement of pit locations and sizes were performed and are shown in Figure 4. This planar map shows the relative location of all pits on the outer surface of the slug and shows the linear positions of the pits in scratches. Representative pits were plotted on the map using coordinates from the grid on the sample. Sizes of the pits were measured and the breached areas of the aluminum clad were calculated. In many cases, the total area of the uranium corrosion was significantly greater than the area of the aluminum opening. This was visually obvious and usually resulted in a blistered or raised area surrounding the hole in the aluminum. The size of the aluminum pits and the total effected area of the uranium corrosion is shown in Table 1. The total breached area of the aluminum was found to be about 3.2 in^2 (20.65 cm^2) with a corresponding area of uranium affected of 7.7 in^2 (49.68 cm^2). With a total cladding surface area of over 172 in^2 (11 cm^2) for the outer slug , the percentage of the uranium surface affected by corrosion is calculated to be about 4.5%.

Metallography and Pit Morphology

Pits on the outer surface of the target slugs were generally classical in nature and most appear to follow the explanation provided in the previous corrosion discussion. Although the proof of hydride attack was not absolute, the location of blisters at some distance from any breached areas indicate a strong possibility that this mechanism, as well as, water attack on the uranium is responsible for the pits that are readily visible. Metallography on sections taken from the slug revealed four types of pits present in the slug. Although, some may look slightly different, most of the pits examined in this investigation looked like those shown in Figure 5. The pit shown in Figure 5(a) is a shallow pit than runs horizontally beneath the clad. The aluminum cladding has been completely corroded away to a size slightly smaller than the affected uranium core. The uranium core has superficial corrosion on the surface and only extends to a depth of 0.04 in.(1.02 mm) The uranium oxide has been dissolved away and no longer is present in this pit except for a very thin layer. The shallow corrosion of the surface of the uranium core may be due to a weak cladding bond to the core in this region allowing some undercutting of the aluminum by the uranium corrosion.

The second type pit is shown by Figure 5(b). This pit is similar to 5(a), but the uranium corrosion has penetrated to a depth of 0.09 in.(2.29 mm). The aluminum cladding has been completely corroded away and the uranium core is completely exposed in this region. Due to the loss of the aluminum cladding, the corrosion product has been washed away and is no longer present. Penetration of the uranium core is deeper in this area because of a strong bonding in the region forcing it's direction downward. Whenever the oxide growth is not strong enough to break the bond of the aluminum /nickel /uranium layer, corrosion appears to continue vertically into the uranium.

Figure 5(c) shows a breach in the aluminum cladding smaller than the affected area of uranium corrosion. Generally, the increased corrosion of the uranium with the aluminum cladding still present will cause a bubble to form around the breach from the expansion of the oxide product. The uranium corrosion has a "V" shaped appearance and the depth of penetration of this pit was found to be 0.12 inches (3.05 mm), the deepest pit found on this slug. Because the aluminum cladding is still present except for the smaller breached area, the corrosion product is still present. Again, the growth of the pit in a vertical position may occur because of a better bond of the aluminum cladding to the uranium in this region. In other regions where the aluminum bonding is not as strong a larger bubble occurs and the corrosion runs in a lateral position like that shown by the pit in Figure 5(d).

Figure 5(d) shows a bubbled area where most of the aluminum cladding is still present. This may have been caused by hydride attack, or could have occurred by a small pinhole breach in the aluminum allowing exposure to water. However, the breach was not large enough to allow the uranium oxide to escape. The uranium oxide caused a 0.09 inch (2.29 mm) bulging of the aluminum cladding with little penetration of the uranium metal. All of the oxide is deposited directly under the aluminum cladding. The corrosion of the uranium continues horizontally with very little penetration into the uranium metal because of a weaker bond in this region.

Figure 6 examines a series of pits which originated in a scratched area of the outer slug surface. This pit has a large opening in the aluminum cladding. As shown by Figure 6(b), there is a large area of uranium exposed, as well as the underside of the aluminum. Because the aluminum cladding is exposed to the basin water on both sides, corrosion occurs on the underside that was bonded to the uranium, as well as the outer surface as shown by Figure 6(c).

Figure 7 shows a large pit located near the end of the slug. Not only was the cladding affected, but also corrosion of the end cap was present. General corrosion of the uranium is present. Corrosion of the aluminum cap is taking place on the outer edge as well as in the region of the cap that is in contact with the uranium core as can be seen in 7(b). Due to a crevice that is in this region, accelerated corrosion may be due to crevice corrosion attack in this area.

Figure 8(a) shows a large pit on the outer surface of the slug. The pit in this area is shown penetrating deeper into the core than most of the pits examined. The depth of this pit may have been influenced by the strength of the cladding bond. A strong bond outside the bubbled region could cause the corrosion of the uranium to penetrate the core instead of extending along the surface between the cladding. When the aluminum cladding is breached and oxide is formed, the uranium oxide holds aggressive ions in this region close to the underside of the aluminum cladding. This may be responsible for the aluminum corrosion on the outer, as well as, the inner surface of the aluminum cladding.

Figure 9 shows a large blistered area near the end of the slug. This corrosion site has an affected area of 1.767 in^2 (11.4 cm^2) but the breach in the aluminum cladding is only 0.047 in^2 (30.32 mm^2). This may be due to a weak bond between the cladding and the core in this region. When the oxide from the corrosion of the uranium grows, the stress pushes the aluminum cladding up forming a bubble. The breached region is not in the middle of the bubble because the bubble grows in the direction of the weakest bond. Corrosion product from the aluminum and the uranium probably mixes in this region and can not escape due the small opening in the aluminum cladding.

Figure 10 represents one area of the slug which contained a number of unusual corrosion sites. These sites show four stages in the development of pits, ultimately, leading to full penetration of the aluminum cladding into the uranium core. In 10(a) a circular shaped grooving in the aluminum is seen. Figure 10 (b) shows similar circular shaped grooving, but a small breach of the cladding is seen in the middle of the circle. Figure 10(c) shows a slightly more advanced stage of uranium corrosion, follow by a total breach of the aluminum leading to full exposure of the uranium core to the basin water as shown in Figure 10(d). The exact mechanism responsible for this unusual corrosion appearance is not known. As the slugs were totally covered by aluminum and uranium oxidation products for several years, the potential exists for oxygen concentration cells to be established, local anodic and cathodic sites to exist, or for crevice corrosion attack to initiate.

The metallography conducted on the Mark 31A slowed that the overall integrity of the slug remains intact. The pitting corrosion on the slugs in the basin continues and as more of the aluminum clad is breached, uranium corrosion will continue to increase with extended storage times.

CONCLUSIONS

The examination of the Mark 31A aluminum clad target slugs removed from the L-Disassembly Basin at SRS showed extensive pitting corrosion. Most of the pitting observed on the of outer surface had breached the clad leading to the formation of voluminous amounts of aluminum oxide, uranium oxide, and other fission product oxide corrosion products. Measurements of cesium-137 activity have confirmed that the corrosion of the fuel and target material in the basin is currently increasing at about 2 dpm/ml per day which is extremely low. The metallograpy on the one unirradiated slug indicated that less than 2% of the cladding on the slug surface is breached by the pitting. This has resulted in less than 5% of the uranium surface being affected by corrosion. The overall integrity of the slug remains intact. As more pitting is initiated on the aluminum and more pits penetrate the clad, the amount of corrosion and activity released to the basin will increase.

Metallograpy of the specimens required development of special techniques in sample preparation. Polishing and grinding were most sensitive to loads applied to the specimens. Most of the work could be done without the use of epoxy to stabilize the oxide, but future work will involve this technique. The use of epoxy is necessary if the objective of the metallography is characterization of the uranium and aluminum grain structure. Without the epoxy, the brittle oxide particles will dislodge from the specimen and contaminate the polishing cloths.

The initiation of pitting appeared to take place, in most cases, where the protective oxide on the aluminum clad was broken by scratches on the outer surface of the target tube. These scratches occur when the slugs are removed from the ribbed outer housing prior to storage in the basin. As the pits grow deeper and larger in diameter, they often join to expose large areas of the uranium metal core to the basin water. This reaction produces the large amounts of corrosion products seen covering the top of the slugs. This process is accelerated by the galvanic couple between the stainless steel slug buckets and the Mark 31A slugs.

Shallow pits and deeper pits were found in the uranium core. Some uranium pits were basically the same size of the breach in the aluminum clad, while in other cases, a significantly larger area of the uranium was affected by corrosion. This expansion of uranium corrosion area may be influenced by the bonding between the clad and the core. A stronger bond restricts the expansion and drives the pitting deeper, whereas, a weaker bond allows the corrosion to proceed laterally, As corrosion develops between the clad and the bond, the oxide product exerts large forces on the aluminum causing it to rupture. In addition, we see the large blisters away from obvious breached areas. These blisters provide evidence that some attack by uranium hydride was probable.

The initiation time for pitting on the aluminum clad of irradiated fuel stored in basin water is likely longer than for an unirradiated element because of the thichness of the oxide coating formed during irradiation. Experience at SRS indicates that the pitting ultimately penetrates the cladding. The pitting morphology of the unirradiated target slugs evaluated in this investigation is expected to be similar to irradiated uranium, but the rate of corrosion and, consequently, the size and depth of the pits will likely be greater.

ACKNOWLEDGMENTS

The information contained in this paper was developed during the course of work under Contract No. DE-AC09-SR18035 with the U. S. Department of Energy. The authors would like to thank W. B. Epling, Jr. and members of the Components Handing Group of the Reactor Department at SRS for providing the target slugs and A.S. Holston and S.B. Coleman for assistance with the metallography.

REFERENCES

1. J. P. Howell, P.E. Zapp, and D.Z. Nelson, "Corrosion of Aluminum Alloys in a Reactor Disassembly Basin," CORROSION/93, paper No. 609, Houston, TX: NACE, 1993).

2. J. P. Howell and P.E. Zapp, "Effect of Water Conductivity on Corrosion of Aluminum Alloys in Spent Fuel Storage," CORROSION/94, paper No. 118, (Houston, Tx: NACE, 1994).

3. C.R. Tipton, Jr., Reactor Handbook, 2nd Edition, p129, Vol. I (1960).

TABLE 1
MEASUREMENTS OF AFFECTED AND BREACHED CLADDING AREAS

X Coordinate (in.)	Y Coordinate (in.)	Area Breached (sq. in.)	Area Affected (sq. in.)
0.5	1.25	none	0.012
0.5	4.5	none	0.196
2	4.5	0.110	0.110
2.75	1	0.313	0.313
2.75	3.5	0.196	0.196
2.75	4	0.049	0.049
2.75	4.25	0.234	0.234
2.75	4.75	0.031	0.156
2.75	5.25	0.234	0.234
2.75	6.5	0.016	0.063
2.75	7	0.442	0.442
3	0.5	0.307	0.307
4.5	4	0.196	0.196
4.5	5	0.110	0.110
4.5	6.5	0.110	0.110
5	2.5	0.049	0.049
5	3	0.110	0.110
5.5	2.5	0.049	0.049
5.5	3	0.110	0.110
5.5	8	0.047	1.767
6	6	0.047	0.785
8.5	0.5	0.281	0.281
10	1.5	0.094	1.767
10	8.3	0.031	0.031
	Total (sq. in.)	**3.169**	**7.681**

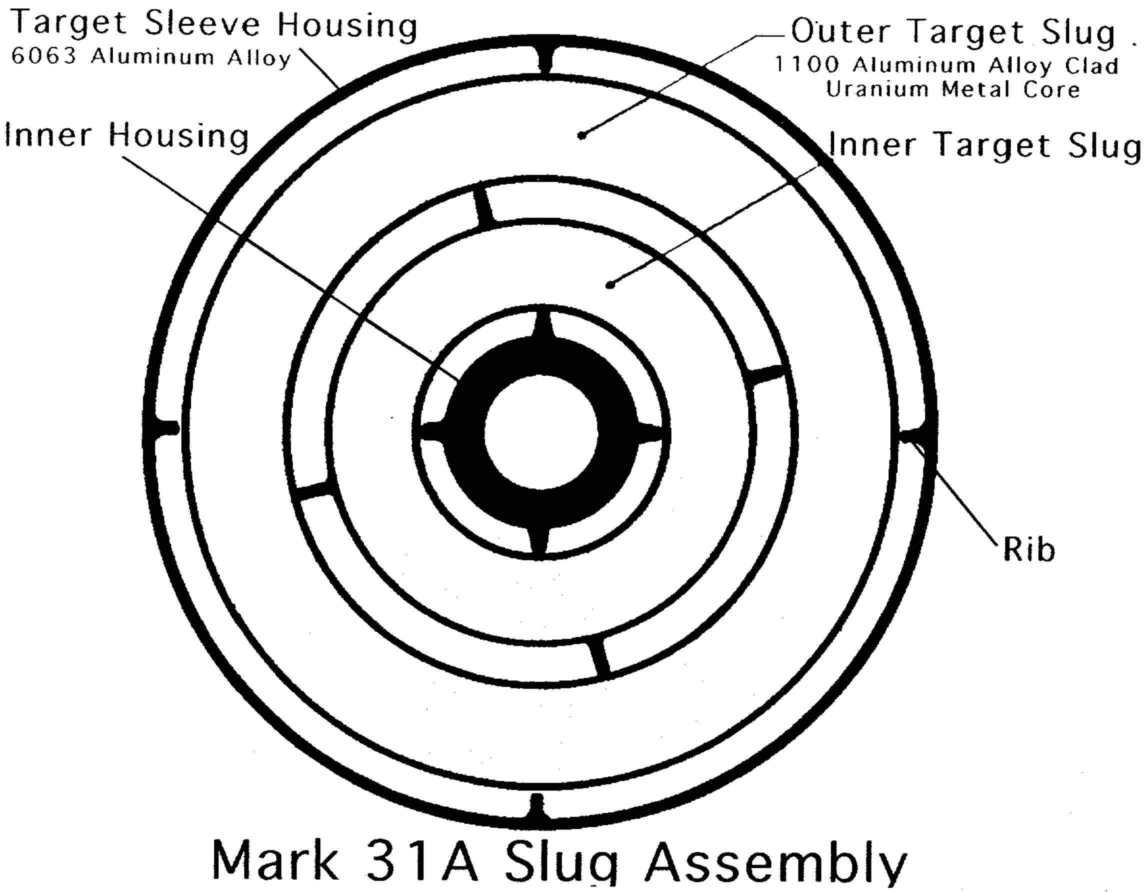

FIGURE 1 - Mark 31A assembly cross section

FIGURE 2 - Stainless steel slug storage bucket

FIGURE 3 - Outer target slug

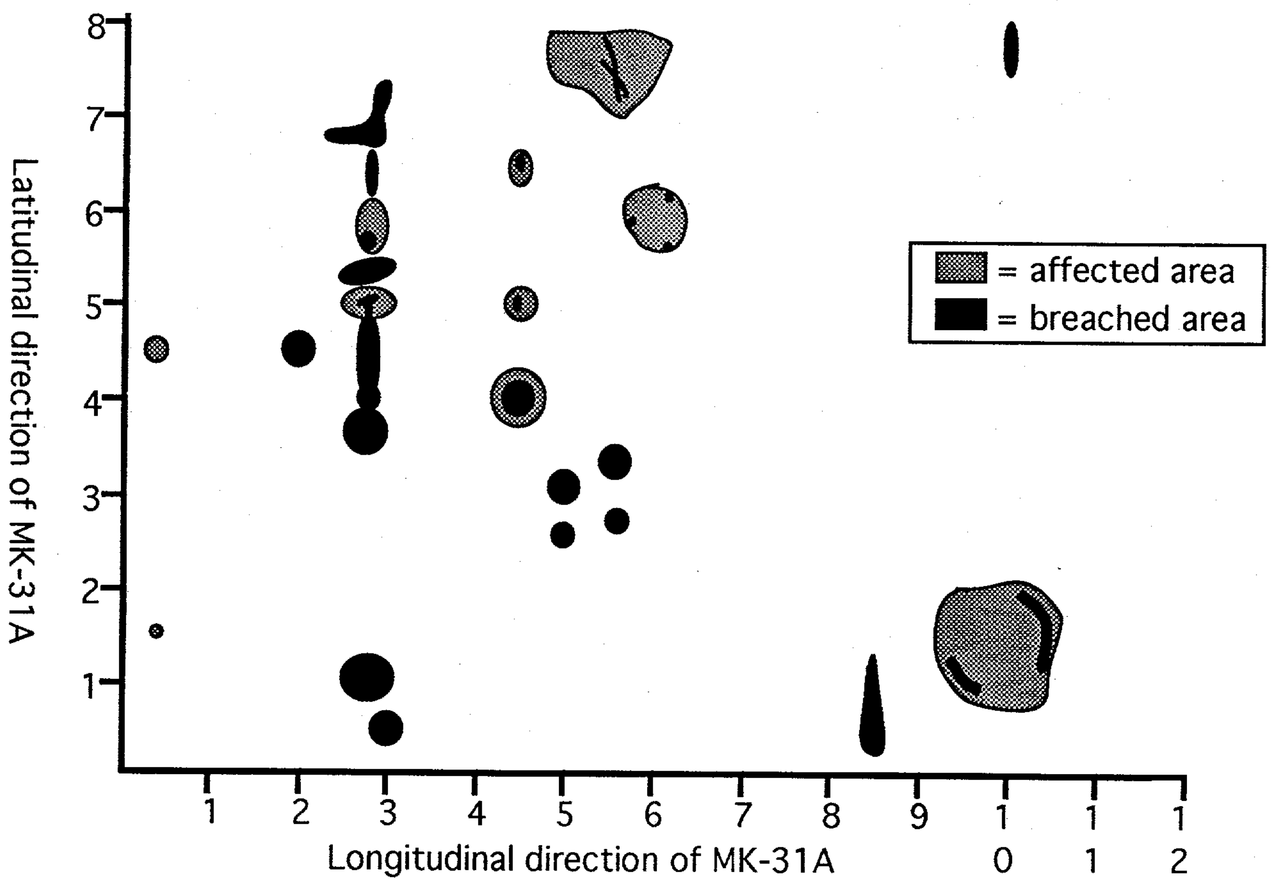

FIGURE 4 - Mapping of corrosion sites on outer target

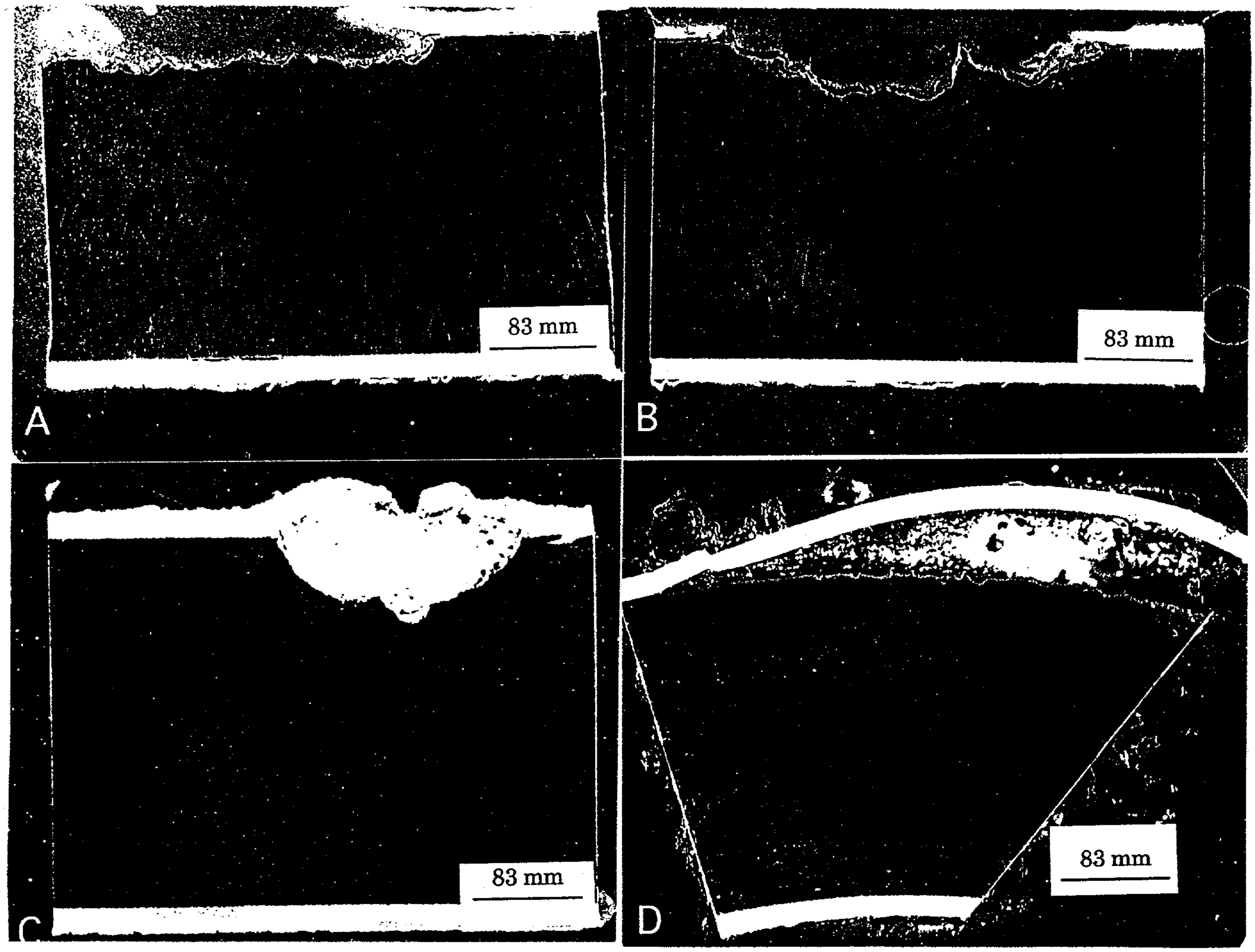

FIGURE 5 - Four typical pit types found on target: (a) shallow corrosion penetration of uranium core, (b) deeper penetration of uranium core, (c) vertical penetration of uranium core, (d) bubble formed under aluminum cladding

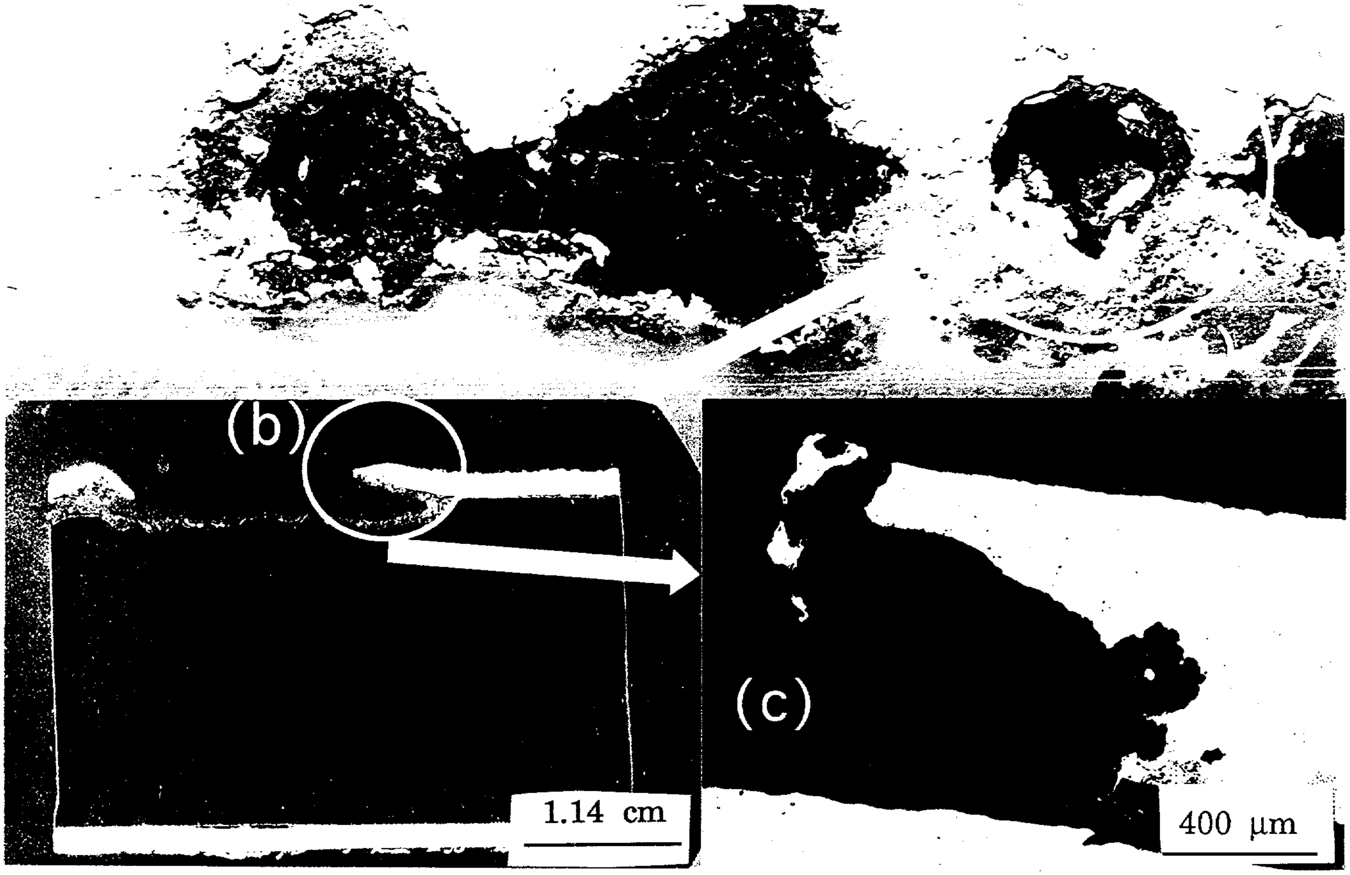

FIGURE 6 - Corrosion morphology of shallow pit

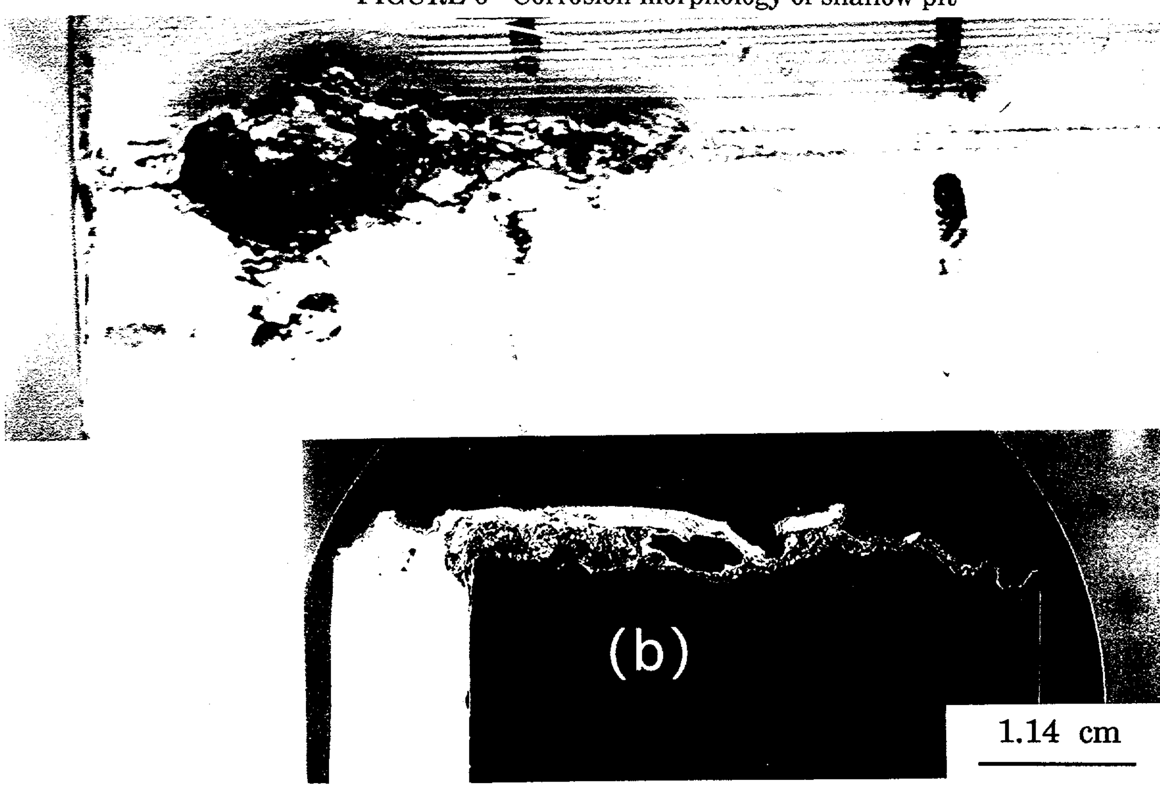

FIGURE 7 - Corrosion pit affecting aluminum end cap

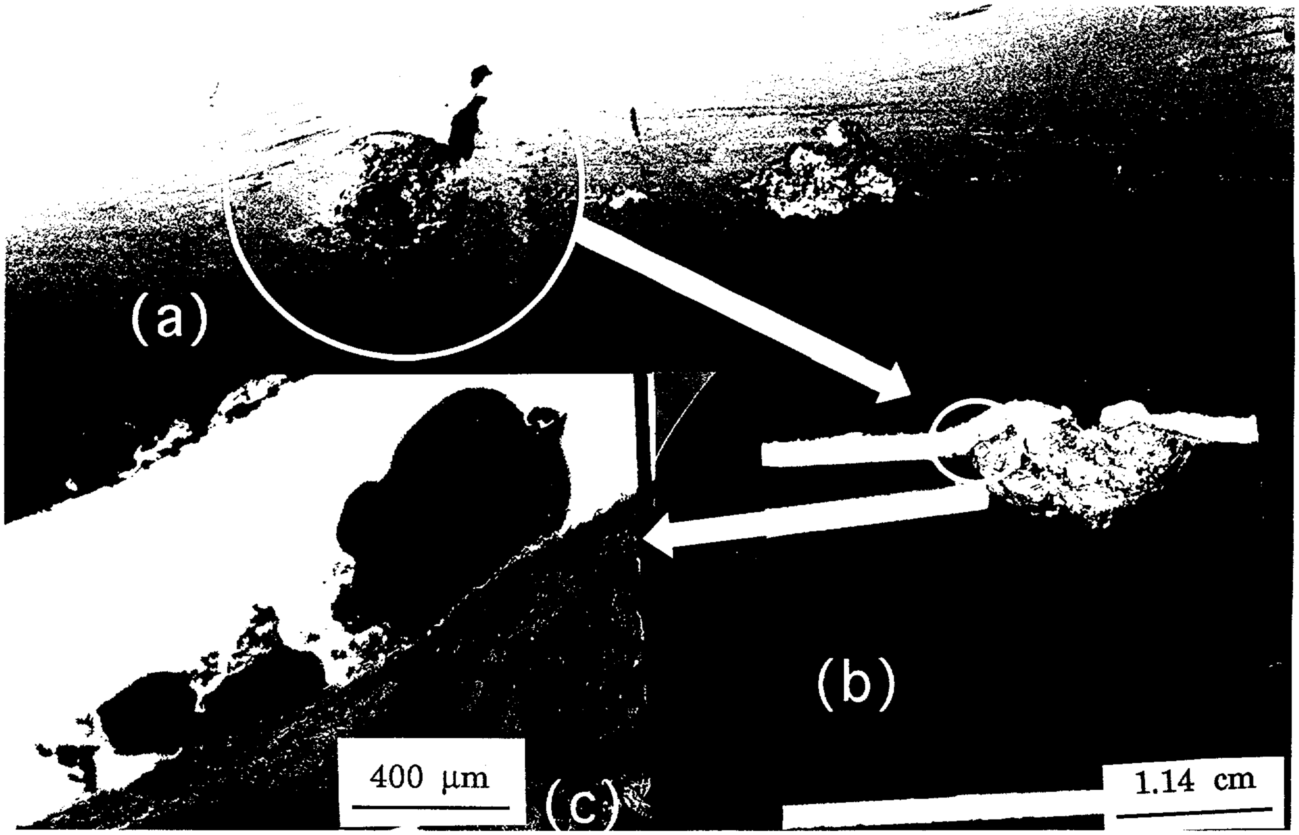

FIGURE 8 - Pit showing bubble and corrosion product still present

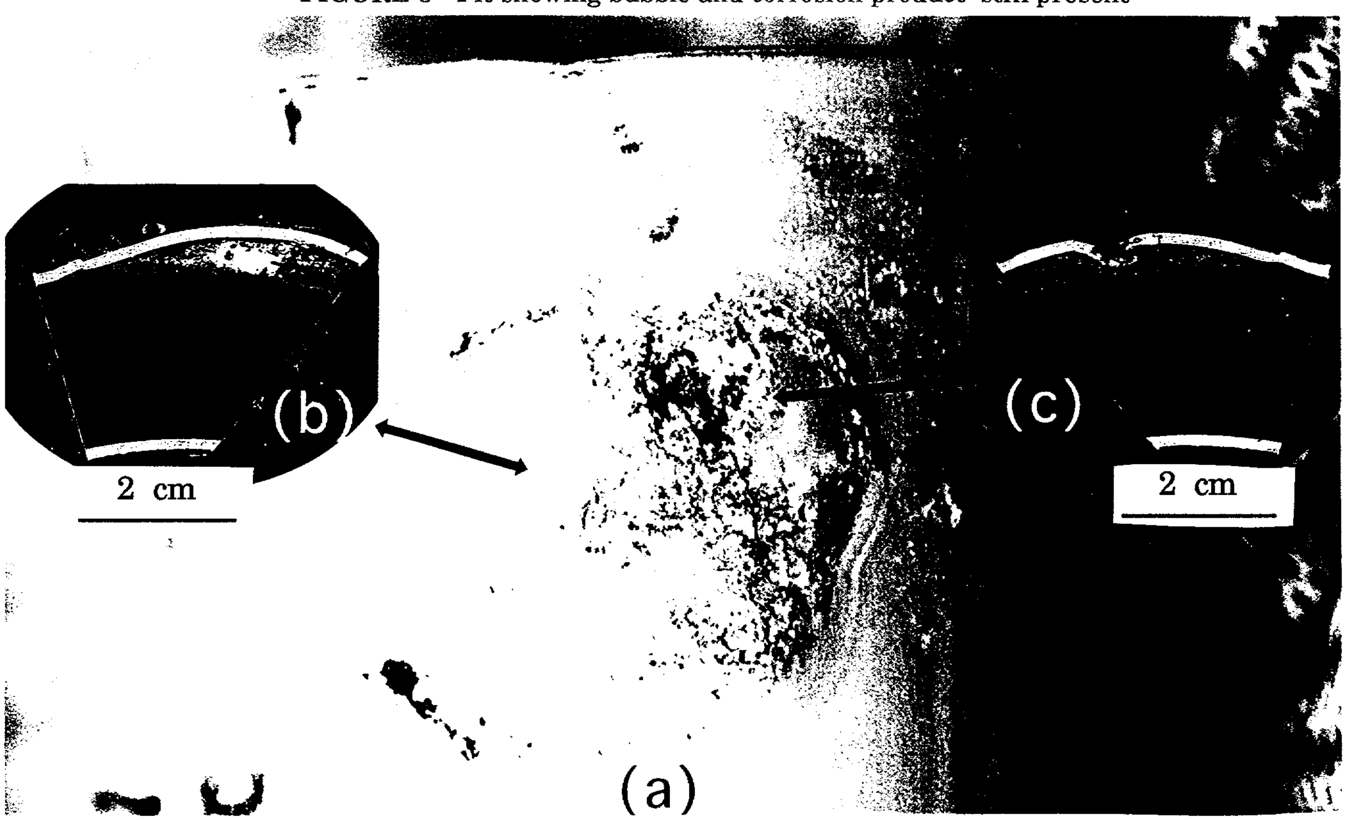

FIGURE 9 - (a) large bubble on target slug, (b) left region of bubble showing debonding, (c) right region of bubble showing debonding

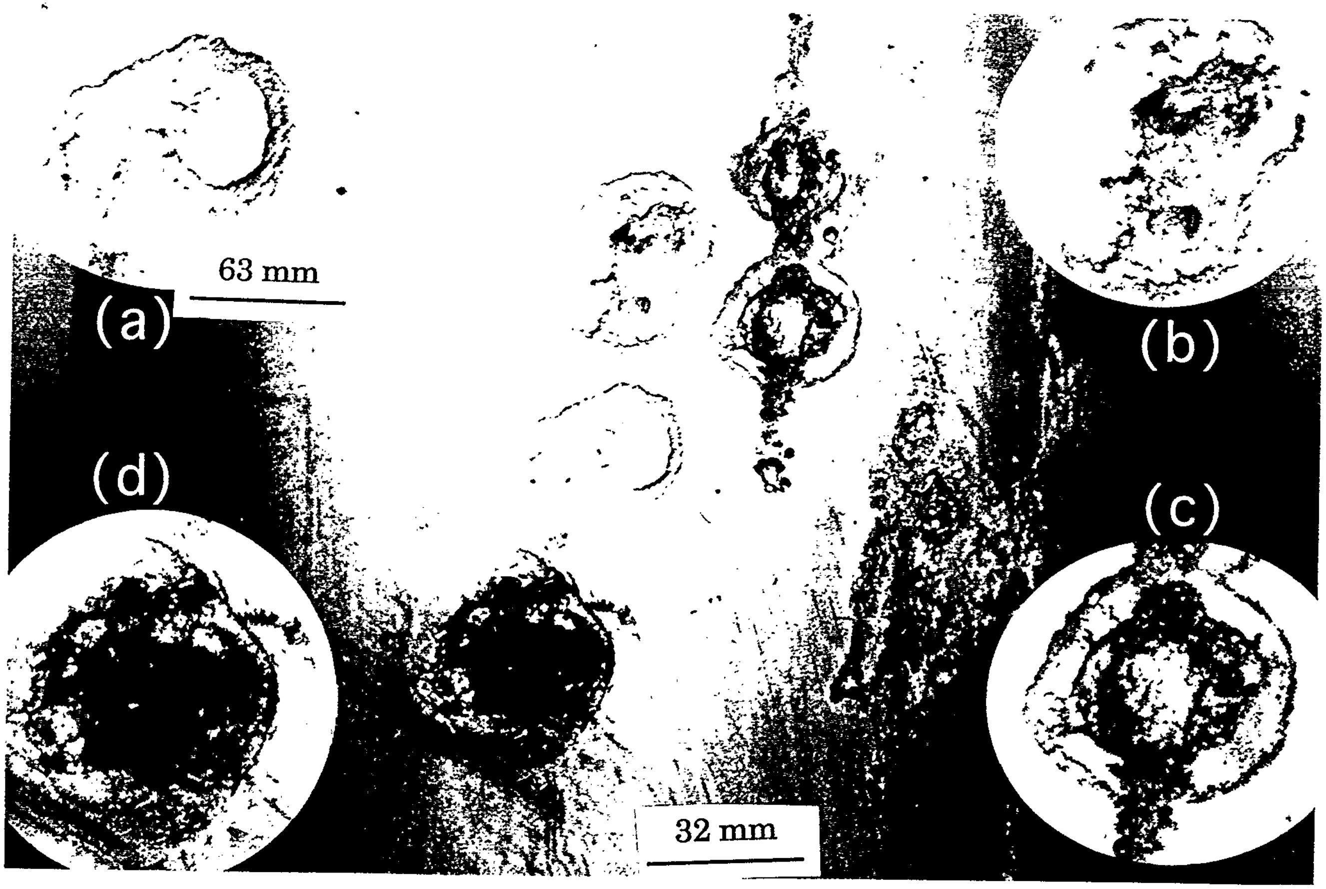

FIGURE 10 - Different corrosion morphology, (a) (b) (c) (d) shows different ages of particular corrosion morphology