Aluminum-Lithium Technology and Savannah River's Contribution to Understanding Hydrogen Effects in Metals

M. R. (Mac) Louthan, Jr.

Abstract

Exposure of aluminum-lithium alloy targets to thermal neutrons in Savannah River Site (SRS) reactors produced tritium, the radioactive isotope of hydrogen that is used in nuclear weapons. The target development program assessed the metallurgical and mechanical properties of aluminum-lithium alloys and determined the factors effecting tritium retention and migration in the target material. An understanding of hydrogen behavior in the target and in other tritium-containing materials was required to assure that tritium-handling operations at SRS were both safe and efficient. The aluminum-lithium and hydrogen-in-metals studies provided the technical basis to assure the combination of tritium retention in target materials during irradiation and transport, successful tritium removal by processing targets in the extraction facility, and successful operation of a tritium packaging and handling systems. The hydrogen-in-metals research programs, which were focused on plant operating needs, also led to discoveries that influenced numerous technologies not generally associated with weapons-materials production. Experimental studies of hydrogen in zirconium and titanium alloys provided a technical basis to avoid stress orientation of hydrides in nuclear fuel claddings and delayed failure of welds on titanium airframes. The susceptibility of austenitic stainless steels to hydrogen embrittlement was demonstrated and broad-based models for hydrogen embrittlement processes were developed. Measurement and analysis of hydrogen uptake and migration kinetics in a variety of engineering materials provided the basis to demonstrate hydrogen transport by dislocations, to identify short circuit diffusion paths in multiphase alloys, and to understand hydrogen trapping at extraordinary sites in a metal lattice. Hydrogen solubility and diffusivity equations were also established for a number of metals. This paper highlights some of the key discoveries associated with the aluminum-lithium and hydrogen-in-metals studies at SRTC.

Introduction

Tritium, an isotope of hydrogen, is a critical component in the construction of thermonuclear weapons. There is very little tritium existing in nature, thus, to support U.S. weapons programs, tritium was produced by the irradiation of lithium in nuclear reactors at SRS. The successful use of tritium to increase the yield of nuclear weapons required a detailed understanding of the behavior of tritium production, extraction, and containment systems. This paper highlights the aluminum-lithium and other hydrogen in metals technologies that emerged from Savannah River during the past four decades of tritium production.

The isotopes of hydrogen are protium, deuterium, and tritium. The nuclei of these isotopes contain one proton and either zero (protium), one (deuterium), or two (tritium) neutrons. All three of the isotopes occur in nature, but most of the existing tritium has been prepared artificially through nuclear reactions such as:

$$_{3}\mathrm{Li}^{6} + {_{0}\mathrm{n}^{1}} = {_{1}\mathrm{H}^{3}} + {_{2}\mathrm{He}^{4}}.$$

This reaction, which depicts the lithium isotope of mass six absorbing a neutron (mass one) and decaying into tritium (mass three) and helium (mass four) atoms, was the basis for tritium production at SRS. Development of the technologies required to safely perform this reaction in a nuclear reactor and to process, contain, and package the resulting tritium was a major SRS contribution to the success of the U. S. nuclear weapons program.

Tritium produced at SRS was mixed with deuterium for use in a nuclear weapon. Such tritium-deuterium mixtures are used to boost the yield of nuclear weapons through thermonuclear reactions such as

$$_1H^3 + {}_1H^2 = {}_2He^4 + {}_0n^1 + 18\ \text{MeV}.$$

The amount of energy released by the fusion of deuterium and tritium atoms to form a helium atom and a neutron is over four million times as great as the amount of energy released by chemical reactions such as

$$C + O_2 = CO_2.$$

The great amount of energy released by the thermonuclear reaction is a primary reason that nuclear weapons can be engineered to have multi-megaton yields. Successfully using tritium to boost the yield of nuclear weapons requires a detailed understanding of the behavior of tritium in the production, extraction, containment, and packaging systems. Tritium must be carefully contained and handled at all stages of processing because it undergoes radioactive decay by emission of a β-particle (an electron) and conversion to a helium atom of mass three.

The metallurgical characterizations of aluminum-lithium alloys at Savannah River, supplemented by national laboratory investigations, provided the technical basis for successful design, fabrication, and irradiation of tritium production targets. This work was one of the first major efforts to fabricate and use standard components manufactured from aluminum-lithium alloys. Many of the property measurements made during the characterization still serve as benchmark references for the aluminum-lithium system. Additionally, the Savannah River efforts to establish foundry practices, to develop fabrication techniques and heat treatment schedules, and to understand material behavior provided part of the experience base required for the aluminum-lithium industry. This industry is now providing aluminum-lithium alloys for aircraft and aerospace components, partially because of the technology and experience developed for tritium production targets.

The broad-based impact of tritium target technology development is even more apparent when the associated hydrogen-in-metals technologies are considered. An understanding of tritium, or hydrogen, uptake and diffusion in metals was required to assure tritium retention in the target during irradiation and storage and tritium release during extraction operations. An understanding of the behavior of hydrogen, including hydrogen effects on the mechanical properties of structural and containment materials, was required for safe and efficient operation of the tritium extraction, packaging, and storage systems. This requirement provided a gateway to large-scale hydrogen-in-metals research because the expertise and facilities necessary for successful tritium operations are identical to those required for solving many other hydrogen-in-metals problems.

Aluminum-Lithium Alloys

Lithium was alloyed with aluminum and processed through the Savannah River fuel and target fabrication facility to form a tritium production target. The targets were then exposed to neutrons (irradiated) in the Savannah River reactors. The irradiated targets were removed from the reactor, allowed to cool in large pools or basins, and ultimately transferred to the extraction facility where tritium was removed, purified, stored, and packaged.

Lithium reacts so readily with oxygen and moisture that it is used as a scavenging/purifying agent for inert gases. This high chemical reactivity prevents the direct exposure of lithium to either air or water. The SRS reactors used heavy water (deuterium oxide) to cool the fuel and target elements and to moderate (slow) the neutrons produced in the fuel elements. In addition, irradiated components are cooled in water-filled storage basins. Therefore, unprotected lithium could not be used as a target material for tritium production. Aluminum was the material selected to protect the lithium and to contain any tritium produced in the target element.

Two protection and containment barriers were established during the fabrication of the aluminum-lithium, tritium production targets. The first barrier against lithium interactions with, or tritium release to, the surrounding environment was the aluminum matrix that surrounded lithium-rich particles in an aluminum-lithium alloy. Predetermined quantities of lithium were placed in an already molten aluminum bath to form this alloy. The melt was then cast to form right circular cylinders of various lengths. These cylinders were covered with relatively pure aluminum and further treated to form slugs that were 2 to 3 centimeters in diameter or tubular elements that were either 4 or 9 centimeters in diameter. The aluminum coverings or claddings for the aluminum-lithium alloys were approximately 0.75 millimeters thick and provided the second protection/containment barrier.

Thousands of target tubes were needed to meet tritium production requirements. Fabrication techniques, alloy specifications, inspection procedures, and irradiation conditions were chosen to assure:

- Reproducible target production to very exacting specifications
- Cladding integrity throughout fabrication, irradiation, and storage
- Tritium containment during irradiation and storage
- Dimensional stability during irradiation
- Efficient tritium release during extraction.

Effective standardization of the processes associated with target fabrication and tritium production required an understanding of the physical and mechanical properties of aluminum-lithium alloys.

Microstructure controls the properties of metals and alloys. Most engineering alloys contain many, very small crystals of various orientations. The individual crystals are termed grains and the size, shape, and arrangement of the grains is part of the materials microstructure. Microstructure is revealed by the examination of specially prepared samples. This examination is termed metallography and generally requires the use of a microscope to reveal the grain structure. The boundaries between grains, precipitates, and inclusions are other examples of microstructural elements in metals. The number, size, shape, and distribution of the microstructural elements can be controlled through metallurgical practices such as heat treatment and deformation. Different production practices will produce different microstructures in the same metal or alloy. Because microstructure plays a major role in controlling the properties of metals and alloys, the properties of a given alloy are significantly influenced by the production practice. The aluminum-lithium alloys used for the tritium production targets contained microstructures that had to be characterized to assure successful production practices. The effects of foundry and fabrication practices on microstructure were determined and techniques to control the microstructure were established. The effects of grain size and precipitate morphology on the mechanical properties were found to be of particular importance.

The metallurgical characterization of precipitate behavior in the target alloys required knowledge of the solubility of lithium in aluminum. The temperature dependence of lithium solubility in aluminum was measured in the early 1960s (Costas and Marshall 1962). This work provided a partial basis for alloy selection and heat treatment practices that increased the room temperature yield strength of aluminum from slightly less than 50 MPa to over 250 MPa. The alloy strength increased with increasing lithium, up to approximately 5 wt% Li, and could be modified through heat treatment. Test samples were extruded and aged (held at an elevated temperature for a prescribed period of time) to develop the high strengths.

Irradiation generates heat at the target interior because of the energy deposition associated with neutron absorption and because of the energy released by the transmutation of lithium to tritium and helium. The heat is removed from the target by reactor coolant flowing across the cladding. This produces a thermal gradient in the target. The size of the gradient and the maximum internal target temperature depend on the amount of heat generated, the temperature of the coolant, the thermal conductivity of both the aluminum-lithium alloy target and the cladding material, and the size of the target. Therefore, the thermal conductivity of the target material needed to be established, and the effects of lithium content and irradiation on thermal conductivity were determined. It was discovered that the temperature gradients in a target during irradiation caused lithium to migrate from hot regions to cold regions (Costas 1962). The rate of migration was dependent on lithium content, temperature, and temperature gradient. Migration was negligible if the temperature was below 473 degrees K or if the lithium content was below 1.5 wt.%. Furthermore, the tendency for lithium migration was also affected by the metallurgical condition of the target material. These were significant observations because lithium migration, which could distort the target, disrupt the coolant flow, and adversely affect reactor operations, could be controlled through alloy selection, heat treatment, and target design.

The effects of alloy selection, fabrication practices, and heat treatment on the mechanical properties of the aluminum-lithium alloy were determined. Tensile properties, hardness, and creep rates were measured, and this knowledge was integrated into the target development processes. Creep, or time-dependent plastic deformation, was of concern because many aluminum alloys tend to creep at temperatures below the anticipated reactor operating temperature. Creep can be caused by atom migration and, in the target alloy, could be controlled by migration of aluminum atoms, lithium atoms, or both. Atom migration, or diffusion, in materials is a thermally activated process that is typically described in terms of the diffusivity. The diffusivity of lithium in aluminum was measured at Savannah River (Costas 1963). The similarity between the temperature dependence for lithium diffusion and for creep in aluminum-lithium alloys demonstrated that lithium atom movement controlled creep. This conclusion was consistent with the observation that, during creep tests, lithium diffused to and precipitated as LiAl on grain boundaries oriented 45-90 degrees to the stress axis (Marshall 1961). This is an example of how microstructure can change when an alloy is exposed to a new environment. The selective precipitation occurred because the density of the LiAl phase, 1.75 gm cm^{-3}, is less than the density of aluminum-lithium matrix, thus the preferential precipitation of LiAl on favorably oriented grain boundaries would selectively expand the alloy along the stress axis. However, lithium redistribution was not observed in tests at temperatures below approximately 473 degrees K, thus providing additional evidence of the importance of temperature control to target stability.

Tritium and Helium in the Target

Studies at Savannah River demonstrated that irradiation changed the metallurgical condition and microstructure of the tritium production targets. Neutron absorption caused the aluminum and lithium atoms in the target to be displaced from their equilibrium positions—termed displacement damage—and produced transmutation products such as tritium, helium, and silicon. Tritium and helium were produced from lithium-6 (Li-6) atoms, and silicon was produced from aluminum. The displacement damage increased the number of disruptions in the alloy, thus increasing the strength and decreasing the ductility and toughness of the material. Silicon accumulations had a similar effect. The level of change in the mechanical properties of the target materials increased with increasing neutron fluence (number of neutrons hitting the target). The amount of tritium and helium generated in the target also increased with increasing neutron fluence. Tritium and helium are relatively insoluble in aluminum and, given the opportunity, agglomerate and precipitate to form gas bubbles in the aluminum alloy matrix. The nucleation and growth of gas bubbles could cause the target to swell during irradiation in much the same fashion as bread rises during baking. Excessive swelling impairs target performance by inhibiting coolant flow, increasing the target temperature, and compromising tritium containment.

The tritium and helium formed in the target behave somewhat differently. The newly formed helium atoms are primarily located in the aluminum matrix while the tritium atoms are distributed in both the matrix and the lithium-rich LiAl phase (Owen and Randall 1976). The tritium atoms react with the LiAl to form lithium tritide and free aluminum by the chemical reaction

$$\mathrm{LiAl} + \mathrm{T} = \mathrm{LiT} + \mathrm{Al}.$$

The equilibrium tritium pressure associated with this reaction is temperature dependent and is given by (Peacock et al. 1995)

$$p_{(\mathrm{equi})} = 351 \exp(-10{,}700/RT) \text{ atm.}$$

This pressure represents the tritium pressure inside a gas bubble or cavity in the target material and, at a reactor operating temperature of 373 degrees K, is only 0.0002 atm if lithium atoms are available for interaction with the newly formed tritium. This pressure is too low to cause target swelling. Additionally, this very low effective pressure provides a minimal driving force for tritium release and thus enhances tritium retention in the target. In the absence of lithium atoms for the reaction, the effective tritium pressure would be given by the relationship

$$p^{1/2} = C/[0.00034 \exp(-15100/RT)] \text{ atm}^{1/2}$$

where C is the tritium concentration, expressed in parts per million (Louthan et al. 1976a). The pressure calculated from this relationship when C is 10 parts per million could not be contained if gas bubbles developed in the target. Therefore, lithium must remain in the target alloy throughout the irradiation in order to prevent tritium-induced swelling and tritium release.

These observations provided the technical basis to specify a maximum Li-6 enrichment of 50%. The Li-7 atoms are not involved in the transmutation reaction and therefore remain in the aluminum-lithium alloy throughout irradiation. This specification assured that even if the target remained in the reactor until all the Li-6 was transmuted to tritium and helium, there would be enough Li-7 remaining in the aluminum-lithium target material to trap (react with) any tritium produced in the target. In addition to preventing tritium induced swelling, trapping greatly reduced tritium migration rates and prevented diffusion-induced tritium losses when irradiating, storing, and transporting targets.

Helium is an inert gas and generally does not interact with other atoms or molecules. Therefore, chemical reactions, similar to the tritium trapping reaction, could not be used to control helium-driven swelling of irradiated targets. This fact, coupled with the success of tritium trapping in preventing tritium-driven swelling, demonstrated that swelling under irradiation is primarily due to helium generation (McDonnell 1989). One helium atom is produced for every tritium atom created by the $^{6}\mathrm{Li}(n,\alpha)^{3}\mathrm{H}$ reaction. Agglomeration of the helium atoms into large gas bubbles would compromise target performance by causing large-scale swelling. Fortunately, the helium atom size is such that, at low helium concentrations, the helium atoms are trapped at microstructural discontinuities in the aluminum-lithium alloy. This type of trapping leads to the formation of small clusters of helium atoms and does not cause appreciable swelling. However, the cluster size grows with irradiation exposure and eventually the build-in of helium will cause the target to swell. The helium concentration required for the onset of swelling is sensitive to the temperature and decreases as the temperature increases.

The swelling threshold at SRS reactor operating temperatures occurs at a helium content of approximately 0.067 wt %. Although this weight fraction looks small, the helium contained in 1 cc of aluminum-lithium alloy at the swelling threshold would occupy approximately 100 cc if released into the air at room temperature. Compression of the 100 cc of helium into a 1-cc volume requires a pressure of 1470 psi. A higher pressure is required to contain the helium in a volume smaller than 1 cc. For example, a pressure of 14,700 psi is required to compress helium into a gas volume of 0.1 cc or 10% of volume occupied by the aluminum-lithium alloy that originally contained the helium. If the temperature is increased from room temperature to reactor operating temperatures for the target, the pressure exceeds 18,000 psi. If only 1% swelling is allowed, the helium pressure at reactor operating temperatures exceeds 180,000 psi. The yield strength of a non-irradiated Al-2wt%Li alloy at 373 degrees K is approximately 14,000 psi and an Al-4wt%Li alloy yields at a stress of approximately 23,000 psi.

The swelling comparisons illustrate the importance of limiting the amount of helium produced in an aluminum-lithium alloy target and demonstrate the necessity to control the chemistry of the target alloy. Integration of the SRS experience base with the accompanying increases in understanding of swelling processes provided the technology to develop the target irradiation limits that precluded excessive swelling during reactor operations.

Extraction of Tritium

The irradiated tritium production targets were removed from the reactor and stored in basins that contain about a million gallons of cooling water. The water flowed around, and through, the tubular targets. Water flow is driven by natural convection and maintains the target temperatures below those experienced during irradiation. Decay of the short-lived transmutation products and activated species lowers the heat-generating capacity of the target tubes, and, after several weeks of basin storage, the targets could be safely transferred to the tritium extraction facility. The entire removal and transfer operations involved remote operations through shielded facilities to minimize personnel exposure and assure operational safety.

Tritium was recovered by placing the irradiated targets in a stainless steel crucible, lowering the crucible-target assembly into an extraction furnace, evacuating the furnace chamber to a pressure below the decomposition pressure of lithium tritide, and heating the evacuated system to promote tritium release. Experimental measurements demonstrated that target swelling and helium release preceded tritium recovery (McDonnell 1989). Target heating promoted the agglomeration of the helium gas bubbles and the formation of interconnected porosity throughout the irradiated aluminum-lithium alloy. This agglomeration caused the target to swell, thus stretching the target cladding. The aluminum cladding then ruptured because its ductility had been reduced significantly by irradiation. Rupture of the cladding exposed the interconnected porosity and released the helium to the vacuum extraction environment. The low extraction pressure caused the lithium hydride, which was surrounded by aluminum, to decompose and release tritium to the extraction furnace. Virtually all the tritium produced during the reactor operation was removed from the target during the extraction operation.

The tritium and helium removed from the target during the extraction operation was collected in stainless steel tanks. The helium is not used in nuclear weapons; thus separation of the tritium from the helium is necessary. To separate the helium from the tritium, the collected gases were passed through a palladium diffuser. The diffuser is essentially a long, thin-walled tube. The gas mixture is passed on one side of the tube while a low pressure is maintained on the other side. The diffuser is heated to promote tritium uptake and diffusion in the tube wall. The combination of uptake and diffusion is termed permeation, and tritium and the other hydrogen isotopes will permeate (pass through) the tube wall. This allows for the collection of tritium, and any other hydrogen isotopes that may be present in the gas stream, on the evacuated side of the diffuser. Helium is virtually insoluble in most metals and alloys, including palladium, and does not permeate the tube. Diffuser operation thus provides a technique to separate the tritium (and other isotopes of hydrogen) from the helium in the extracted gas stream. However, all three hydrogen isotopes permeate the diffuser and the collected "tritium" gas stream also contains protium and deuterium. Protium, deuterium, and tritium separated from each other by cryogenic distillation and the final product is high-purity tritium. The high-purity tritium is packaged and stored for use in atomic weapons.

Associated Hydrogen-in-Metals Technologies

Advances in aluminum-lithium technology for tritium production at the Savannah River Site required the development of a large number of associated hydrogen-in-metals technologies. An understanding of tritium, or hydrogen, uptake and diffusion in metals was required to assure tritium retention in the target during irradiation and storage and tritium release during the extraction operation. The behavior of hydrogen, including hydrogen effects on the mechanical properties, in structural and containment materials was required for safe and efficient operation of the extraction, packaging, and storage systems. The expertise and facilities required for successful tritium production operations are identical to those required for solving most other hydrogen-in-metals problems. Therefore, the technical support system for the aluminum-lithium technology program also enhanced a variety of other hydrogen technologies not directly related to tritium production. The Savannah River work contributed significantly to the:

- Prevention of hydride-induced failure of zirconium alloys used in the nuclear power industry
- Avoidance of cracking during welding of titanium alloys of interest to NASA for supersonic aircraft
- Determinations of hydrogen solubility, diffusivity, and permeability in, and measurement of the effect of hydrogen on the mechanical properties of, infrastructure materials required to support a hydrogen economy
- Development of the fundamental aspects of hydrogen-metal interactions in a variety of metals and alloys
- Demonstration that helium, introduced through radioactive decay of tritium, increased the strength and decreased the ductility and weldability of tritium exposed metals
- Determination of the effect of helium on hydrogen-induced slow crack growth in austenitic stainless steels
- Discovery that helium implanted during irradiation can cause cracking during weld repair of nuclear reactor components.

The last three items listed have direct application to the selection and use of metals and alloys in fusion reactors and in advanced spallation neutron sources.

Hydride Cracking of Zirconium Alloys

Zirconium-based alloys are used for fuel cladding, process tubes, and structural elements in the nuclear power industry. These alloys have a strong affinity for hydrogen and, when sufficient hydrogen is absorbed, will precipitate hydride phases throughout the zirconium microstructure. These hydride precipitates can have deleterious effects on the mechanical properties of zirconium alloys. In the early 1960s, emerging experimental data suggested that the tolerance of zirconium alloys for hydrides was rather high and early concern over the potential for hydrogen embrittlement was subsiding. However, experiments at SRS demonstrated that the influence of hydrogen on the mechanical properties of Zircaloy (a zirconium-based alloy that contains small amounts of tin, iron, and nickel) was determined by the orientation of the hydride platelets. Small amounts of hydrides could have very deleterious effects on the mechanical properties if the platelets were oriented perpendicular to the direction of applied stress (Caskey et al. 1961; Louthan and Marshall 1963; Marshall and Louthan 1963). The orientation of the platelets was controlled by stresses in the Zircaloy during hydride precipitation. Platelets tended to precipitate with their broad faces perpendicular to tensile stresses and parallel to compressive stresses. This tendency was termed stress orientation (Marshall and Louthan 1963).

Zirconium alloys have a hexagonal-close-packed (hcp) crystal structure. Plastic deformation in hcp structures is anisotropic. This anisotropy leads to the development of crystalline textures or preferred orientations in most wrought products. The nature of the preferred orientation in any given component depends on the fabrication process used to manufacture that component. Measurements of the extent of stress orientation in tube sections with various preferred orientations (Marshall 1967) confirmed that the orientation of the hydrides in Zircaloy, and the susceptibility of Zircaloy to stress orientation were controlled by the preferred orientation of the Zircaloy matrix. Therefore, the processing technique use to manufacture the Zircaloy cladding, tube, or component had a major influence on the tendency of the materials to undergo stress orientation (Louthan and Marshall 1963; Marshall 1967). This observation demonstrated that a potentially significant problem with the use of Zircaloy as a nuclear fuel cladding could be mitigated through the selection of manufacturing technologies to control the preferred orientation of the material. These results provided the foundation for the technical basis that currently assures against adverse hydride orientations in fuel claddings and process tubing used in commercial power reactors.

Hydrogen in Titanium Alloys

The Savannah River observations that lithium diffused to and precipitated as LiAl on grain boundaries oriented 45 to 90 degrees to a tensile stress, and stresses caused preferential hydride orientations in Zircaloy led to the discovery that titanium alloys were also susceptible to stress orientation of hydrides (Louthan 1963). This observation was one of the keys to understanding delayed failure in titanium alloys.

Hydrogen uptake may occur in service or be introduced by fabrication processes such as welding. Titanium alloys react with moisture in the environment and release atomic hydrogen at the metal surface. This hydrogen may move through (permeate) the titanium and accumulate in regions of high tensile stress. The high stresses may be the result of service loads or may be residual stresses introduced by fabrication and/or assembly processes. When sufficient hydrogen has accumulated in the high stress area, a hydride phase will precipitate and will be oriented so that the broad face of the platelet is perpendicular to the tensile stress. This orientation is favorable for crack nucleation and/or growth along the hydride/metal interface or through the hydride itself. Once the hydride has cracked, the high stresses, which promoted hydrogen accumulation, will now be just beyond the crack tip. The accumulation, precipitation, cracking sequence will then be repeated.

The hydrogen accumulation and hydride precipitation processes are both diffusion controlled and require the passage of time. Under these conditions, hydrogen-induced cracking may initiate sometime after a crack-free part has been placed in service. The delay time for cracking can vary from hours to years, depending on the hydrogen content, stress level, microstructure of the titanium alloy, and temperature of exposure. The publication of observations on stress orientation of hydrides in titanium caused multiple interactions with National Aeronautics and Space Administration (NASA) because of the use of titanium in airplanes and aerospace applications. The continued importance of these results is apparent in the 1993 edition of the *Metals Handbook* (Metals Handbook 1993), which describes hydrogen-induced delayed cracking of titanium welds as resulting from the time-dependent precipitation of stress oriented hydrides in the high tensile stress regions of the welds.

The National Aeronautics and Space Administration sponsored several experimental programs at Savannah River, including an investigation of hot salt stress corrosion cracking of titanium alloys. The research, conducted in the mid to late 1960s, demonstrated the hot salt stress corrosion cracking process involved two distinct steps: crack initiation and crack propagation (Rideout et al. 1969; Rideout et al. 1970). Crack initiation occurs after an incubation period that depends on the temperature of exposure, chemical composition of the salt deposit, and composition of the titanium alloy. The presence of moisture in the salt deposit and the formation of HCl gas by hydrolysis also play vital roles in the cracking sequence. Radiotracer studies, using tritium as a tracer, showed that corrosion produced hydrogen is absorbed by the metal. It was proposed that the absorbed hydrogen accumulated in regions of high, localized tensile stresses. Cracks initiated when the hydrogen concentration exceeded some critical level. Crack propagation was found to be less sensitive to temperature than crack initiation. The stress level and the nature of the salt deposit controlled the rate of crack propagation. Crack propagation occurred primarily by mechanical rupture processes and little or no evidence of corrosion was found on the fracture surfaces. The importance of these observations to understanding and mitigating hot-salt stress corrosion cracking in titanium alloys was emphasized in an article published in the *ASM Source Book on Titanium and Titanium Alloys* (Petersen 1982).

Hydrogen Compatibility

The "Hydrogen Economy" emerged in the 1970s as a technology of importance to the future. This emergence was primarily because of the long lines at gas pumps but also as part of the ongoing quest for clean energy technologies to replace burning fossil fuels. The Energy Research and Development Administration (ERDA), the predecessor organization to the Department of Energy, established an Interagency Agreement through which NASA provided assistance to ERDA's hydrogen energy storage program. The production, storage, transportation, and use of hydrogen as an energy carrier were investigated through this cooperative research and development program and through associated programs conducted at various ERDA sites, such as Savannah River. The Savannah River contributions to the "Hydrogen Economy" technologies were determinations of the compatibility of metals and alloys with hydrogen environments.

Compatibility studies included the determination of hydrogen uptake and migration kinetics in, and measurements of hydrogen effects on the mechanical properties of, structural materials. The Savannah River hydrogen-in-metals technologies, developed to support the production, extraction, storage, and packaging of hydrogen isotopes for nuclear weapons were identical to the hydrogen-in-metals technologies required to support the hydrogen economy.

Data were necessary to assure satisfactory hydrogen containment in hydrogen storage vessels, gas transfer lines, valves, pumps, and the associated storage and transfer equipment. Satisfactory containment included assuring that potential losses of hydrogen because of uptake and/or permeation were minimized and that the materials of construction were compatible with high-pressure hydrogen environments. Savannah River had investigated the behavior of approximately 50 commercial alloys in hydrogen environments by the mid 1970s (Louthan and Caskey 1976a).

Radioactivity provides a significant tool that enhances the detectability of tritium. This enhanced detectability allowed the measurement of the behavior of hydrogen in metals under conditions not previously accessible to measurement. The diffusivity and permeability of hydrogen in cobalt (Caskey et al. 1974), nickel (Louthan et al. 1975a), molybdenum (Caskey 1975), copper (Caskey et al. 1976), aluminum (Louthan et al. 1976a), and titanium alloys (Louthan et al. 1975b) were measured at near ambient temperatures. Hydrogen diffusivity, solubility, and permeability were also measured in austenitic and ferritic/martensitic steels (Louthan and Caskey 1976; Louthan and Derrick 1975; Caskey and Derrick 1974; Louthan et al. 1976b). The data and analysis from the studies provided the technical basis to assess hydrogen uptake and migration in many of the commercial alloys that were being considered for use in the hydrogen production, storage, and distribution systems required if the U.S. were to move toward a "hydrogen economy". The studies demonstrated that hydrogen trapping at metallurgical discontinuities (dislocations, grain boundaries, phase boundaries, alloy and impurity atoms, etc.) played a major role in hydrogen uptake and migration (Louthan et al. 1975a; Caskey et al. 1976; Caskey and Allinger 1974; Louthan 1974) and that surface films, normally present on these commercial alloys, play a major role in hydrogen absorption and permeation processes (Louthan and Caskey 1976; Louthan et al. 1975b; Louthan and Derrick 1975). Generally, the highest permeabilities to hydrogen were found in the iron-based alloys. The transition metals such as nickel and cobalt also showed high permeabilities to hydrogen. Aluminum and copper alloys showed the lowest permeabilities, partially because the hydrogen solubility in these metals is so low.

The room-temperature tensile properties of these same commercial alloys were measured before, during, and after exposure to high-pressure hydrogen environments (Louthan and Caskey 1976; Louthan 1974; Louthan et al. 1972; Capeletti and Louthan 1977). These studies demonstrated that all alloys show some evidence of susceptibility to hydrogen embrittlement. The aluminum alloys were the most resistant to hydrogen-induced degradation. Copper alloys also displayed excellent resistance to hydrogen damage. Iron-, nickel- and titanium-based alloys were all very susceptible to hydrogen damage; however, the extent of damage was dependent on pre-test exposure conditions, sample surface finish, hydrogen pressure during testing, and the metallurgical condition of the alloy. The data and analyses developed from these Savannah River investigations continue to provide a significant part of the technical basis for selection and use of alloys for service in hydrogen environments. This is particularly true for austenitic stainless steels because of a handbook published by Savannah River (Caskey 1983).

Fundamental Aspects of Hydrogen-Metal Interactions

The development of basic data necessary to assure the compatibility of metals and alloys with hydrogen was accompanied by an increased understanding of the fundamental aspects of hydrogen-metal interactions. The Savannah River data and analysis demonstrated that absorption and permeation of hydrogen, as well as the final distribution of hydrogen throughout a metal structure, could not be predicted accurately by the usual thermodynamic and diffusion relationships. Surface effects (particularly the properties and stability of oxide films), trapping by impurity and microstructural defects (discontinuities), and hydrogen transport by moving dislocations all profoundly influence hydrogen motion and distribution (Louthan and Caskey 1976). These three factors also effect hydrogen embrittlement because the quantity and distribution of absorbed hydrogen are directly related to the hydrogen embrittlement mechanism. Because of the importance of trapping to the understanding of hydrogen embrittlement processes, finite-difference techniques were developed to analyze the trapping process (Caskey and Pillinger 1975).

Tensile tests of stainless steel and nickel in high-pressure tritium environments (Louthan et al. 1972; Donovan 1976) demonstrated that plastic deformation had profound effects on the absorption and subsequent distribution of tritium in the test material. Localized, high tritium concentrations were identified by autoradiographic techniques (Louthan et al. 1972). Tensile and/or bend tests with tritium charged iron, Type 304L stainless steel, Alloy 718, and Type 5086 aluminum demonstrated the tritium was associated with dislocations and moved when the dislocations moved (Louthen et al. 1972; Donovan 1977). Dislocations are microstructural defects that are found in virtually all metals and alloys. Dislocation motion is generally responsible for plastic deformation in metallic structures. The observations that hydrogen attached to and moved with dislocations and that such motion can lead to localized, high hydrogen concentrations are two of the basic precepts for hydrogen embrittlement in metals and alloys that do not form hydrides or other hydrogen-rich phases. The Savannah River paper that initially presented these observations was republished in the ASM International book, *Hydrogen Damage*. This book collected, from the thousands of papers discussing hydrogen-in-metals, 30 "key contributions to the understanding of hydrogen damage and to our efforts to overcome the problems it continues to bring" (Hydrogen Damage 1975).

Autoradiographic studies demonstrated that hydrogen diffusion in rutile is anisotropic (Caskey 1974a). Rutile is titanium dioxide and forms naturally on the surface of titanium alloys exposed to air, water, and/or other oxygen containing environments. Oxide film formation on titanium is strongly influenced by the orientation of the underlying metal because the oxide forms in specific orientations with respect to the underlying metal. The oxide-metal epitaxy and the anisotropy in hydrogen diffusion were used to explain hydriding processes in titanium alloys used in hydrogen service (Caskey 1974b).

Other Savannah River studies related to fundamental aspects of hydrogen-in-metals include finding that:

- Isotopic effects on hydrogen transport in nickel are predictable from absolute rate theory if hydrogen acts as an anharmonic oscillator (Louthan et al. 1974a).
- Hydrogen expands the lattice and lowers the stacking fault energy of austenitic steels (Holzworth and Louthan 1968).
- Hydrogen lowers the cohesive energy of coherent twin and grain boundaries (Capeletti and Louthan 1977; Caskey 1983).
- The heat of solution for hydrogen is related to the electron density of states at the Fermi surface (Louthan et al. 1972).
- Hydrogen increases the lattice friction stress of austenitic stainless steels (Caskey 1983).
- Hydrogen can either suppress or enhance the formation of strain-induced martensite in austenitic stainless steels depending on the composition of the steel and temperature (Caskey 1983).
- High hydrogen solubility, low stacking fault energies, and high yield strength (Louthan et al. 1972) promote hydrogen embrittlement.

These fundamental observations provided a technical basis for a phenomenological model for hydrogen embrittlement (Louthan 1987).

Tritium Decay and Helium Accumulations

The radioactive decay of tritium is by the $^3H(b)^3He$ reaction. The helium (^3He), in a tritium-charged metal or alloy, is "born" in the metal lattice. Normally, helium does not dissolve in metals, but the in-lattice birth places a helium atom in solid solution in the metal and can lead to a significant helium build-in when tritium charged samples are stored for long periods of time. Helium atoms do not "fit" in either the normal lattice sites or the interstitial sites where hydrogen atoms generally reside. Helium embrittlement of metals is well known in the nuclear power industry because helium atoms are transmutation products in many nuclear reactions. This embrittlement is considered to be an elevated temperature process and was observed in tritium charged Type 309 stainless steel in the mid 1970s (Louthan et al. 1976c). These initial studies showed that helium build-in increased the strength of the steel but did not significantly effect the ductility until the helium-containing material was heated for a half-hour at 973oK. Subsequent studies demonstrated that elevated temperatures were not requiref for embritvlemgnt (Rawl et al. 1980; West and Rawl 1980). Austenitic stainless steel tensile bars that were tritium charged and aged until the helium concentration exceeded 200 appm and then tested at room temperature, failed along grain boundaries. The ductility was not restored when the samples were vacuum outgassed to remove the remaining tritium (West and Rawl 1980).

Room temperature helium embrittlement was not anticipated because helium is highly trapped. The elevated-temperature helium embrittlement studies suggested that the helium-induced failures result from the accumulction of helium bubbles along grain boundaries (Louthan 1976c). This failure mechanism requires helium migration to grain boundaries, which should be minimal at or near room temperature. Studies with iron and austenitic stainless steel confirmed the intergranular failure process and showed that the formation of helium bubbles was accelerated by a tensile stress (Donovan 1980). These observations demonstrated that helium-induced effects on the mechanical properties had to be evaluated before the long-term safety of tritium containment systems could be assured. Additionally, these results had significant implications to the weldability of irradiated metals and alloys.

These implications became apparent during attempts to repair the Savannah River C-Reactor tank by welding (Kanne 1988). Stress corrosion cracks in a curved transition piece that connected the tank sidewall to the bottom of C Reactor caused the reactor tank to leak in the late 1960s. The reactor was shut down, repaired (using remote gas tungsten arc welding techniques), and returned to service. The tank leaked again in 1984, and a program was initiated to repair the new leaks. Placement of patches over the cracks was, as in 1968, determined to be the best method of repair (Kanne 1988). A robotic-operated service arm was designed and built. Approximately 20 types of end effectors, including those for repair welding, were included in the design. A segmented patch was welded in place and then bubble tested by pressurizing the space between the tank wall and the patch with gas, raising the water level inside the tank, and monitoring for bubbles. Several leak sites were found. Subsequent, dye-penetrant testing showed toe cracking in the welds on the reactor wall (Kanne 1988). The tank wall contained approximately 3 appm of helium, which was introduced into the stainless steel by irradiation-induced transmutations of the alloy element nickel and the impurity element boron. An extensive test program demonstrated that the toe cracks were caused by the welding-induced agglomeration of helium bubbles along the grain boundaries. Test welds in materials that were charged with helium by the radioactive decay of absorbed tritium played an integral role in proving that the weld toe cracking was caused by the presence of helium (Kanne 1988).

Demonstration that small quantities of helium dramatically reduced the weldability of irradiated stainless steel had major implications on the design and repair of components and systems for reactors, accelerators, and other systems where helium implantation will accompany service. Additionally, the use of tritium charged and aged samples to simulate irradiation-induced helium became a standard technique to evaluate techniques for weld repair of irradiated materials. The 1988 discovery of helium effects on the weldability of metals and alloys provided the technical basis for several other Savannah River programs to validate weld repair techniques, quantify the level of helium required to cause weld cracking for different weld techniques, and model the weld cracking processes. One of the current programs is focussed on applications to fusion energy systems.

Hydrogen, Helium, and Slow Crack Growth

Tensile tests of hydrogen-charged austenitic stainless steels demonstrated that surface cracking accompanied plastic deformation. Metallographic examination of failed samples showed that hydrogen accumulation changed the fracture mode from a ductile to a brittle failure process. The brittle fracture modes included grain boundary and twin boundary cracking as well as cleavage fracture. The observation of hydrogen-induced brittle fracture modes raised concerns over hydrogen-induced slow crack growth in austenitic steel systems and components used for hydrogen containment (Caskey 1983). Slow crack growth could cause delayed failure hydrogen embrittlement and lead to sudden failure of in-service components. Tensile tests, which dominated the Savannah River hydrogen-in-metals studies throughout the 1970s, provided virtually no information that was relevant to slow crack growth. Therefore, by the mid 1980s, the hydrogen-in-metals test program had evolved to a focus on fracture mechanics studies to provide crack growth and fracture toughness data (Caskey 1983).

The fracture toughness of a material is generally expressed in terms of a stress intensity factor. This factor provides a measure of the level to which a crack or other sharp flaw enhances the effect of an applied stress. Values for the stress intensity factor depend on the type and magnitude of the load applied to the material and the size of the crack in the material. Failure takes place when the stress intensity factor reaches a critical level. This critical level is termed the fracture toughness of the material. Work at Savannah River demonstrated that tritium, and its decay product helium, lower the fracture toughness of austenitic stainless steels (Caskey 1983; Morgan and Tosten 1990). The extent of lowering depends on the strength (Caskey 1983) and metallurgical condition of the steel (Morgan and Tosten 1990). Other Savannah River studies demonstrated that hydrogen and helium could also cause slow crack growth in austenitic stainless steels that were loaded to stress intensities that were less than the critical stress intensity (Morgan and Lohmeier 1990). Subsequent, fracture mechanics type studies (Morgan and Tosten 1996) demonstrated that:

- Tritium exposed and aged steels had lower fracture-toughness values and less resistance to crack growth than unexposed steels.
- Fracture toughness values were reduced further as the concentration of helium increased.
- The tendency toward intergranular fracture increased as the concentration of helium increased.

Tensile testing demonstrated that, in addition to lowering the fracture toughness and the resistance to crack growth, both hydrogen and helium increased the strength and decreased the ductility of these steels (Morgan 1991). The helium-induced increases in strength enhance the effects of hydrogen on fracture toughness and crack growth. These data provided the technical basis to model slow crack growth in tritium containing stainless steels (Morgan 1997; Morgan 1999).

The model (Morgan 1999) shows that tritium is absorbed into the container wall and migrates through the wall by diffusion controlled processes. Helium then builds in from decay of the absorbed tritium. The helium build-in strengthens the metal but also lowers its resistance to crack growth. If the applied and residual stresses are high enough, a crack can nucleate and grow through the weakened or embrittled region. In this model, the crack could continue to grow in a stepwise fashion:

- Tritium diffusion and decay to helium embrittles the area near the crack tip.
- The crack then propagates through the embrittled zone.
- The crack arrests when it has propagated into non-embrittled material.
- Tritium absorption, diffusion, and decay begin to embrittle the new, near crack tip region.

As the crack grows through the material, the stress intensity level may increase (depending on the geometry and loading conditions). If this is the case, eventually, the non-embrittled region will be stressed to a level where crack growth can be driven by the presence of tritium. When this occurs, the material will fracture. This model—coupled with the Savannah River measurements of crack growth rates, critical stress intensity, factors and the effects of hydrogen and helium on critical stress intensity factors—supports the technical basis to assure the safety of tritium storage containers packaged and handled at Savannah River.

The observation that hydrogen and helium lower the fracture toughness and increase crack growth rates in stainless steels has significant implications to the behavior of metals and alloys in fusion and accelerator driven systems. The high-energy particle beams associated with these emerging energy systems will cause significant transmutations in the exposed areas. High concentrations of hydrogen and helium will therefore accumulate, decreasing the fracture toughness of the structural material. This decrease must be included in the design criteria for such systems.

Conclusion

The aluminum-lithium technology development at Savannah River, especially including hydrogen isotope and helium effects, has made significant scientific and technological contributions to the materials/metallurgical communities. These contributions extend far beyond the nuclear weapons materials production arena. In many respects, the contributions have favorably impacted the every day lives of most U.S. citizens and have the potential to continue to impact our society well into the 21st century.

Acknowledgment

The work described in this paper includes contributions from numerous engineers, scientists, and managers. The net result of these contributions is a chain of achievement linked together through the efforts of individuals. Unfortunately, listing all the individuals is impractical and listing only a few is unfair. Many of the technology developments sited were headquartered in materials oriented groups as the Savannah River Laboratory and the Savannah River Technology Center. The leadership of those groups has been outstanding and included: Phil Permar, Dick Huntoon, Jim Stone, Tami Capeletti, and Natraj Iyer. These leaders are thanked for providing a work atmosphere that fostered high-quality, cooperative research and development, and each individual contributor is thanked for being an integral part of that atmosphere and of the resulting accomplishments.

References

Capeletti, T. L., and M. R. Louthan, Jr., 1977, "The Tensile Ductility of Austenitic Stainless Steels in Air and Hydrogen", p. 153, *Journal of Engineering Materials and Technology*, Vol. 99.

Caskey, G. R., Jr., 1974a, "Diffusion of Tritium in Rutile", p. 109*, Material Science and Engineering*, Vol. 14.

Caskey, G. R., Jr., 1974b, "The Influence of a Surface Oxide Film of the Hydriding of Titanium", p.465 *Hydrogen in Metals*, American Society for Metals, Metals Park, Ohio.

Caskey, G. R., Jr., 1983, *Hydrogen Compatibility Handbook for Stainless Steels*, DP-1643, E. I. DuPont de Nemours & Co., Savannah River Laboratory, Aiken, SC (June 1983).

Caskey, G. R., Jr., A. H. Dexter, M. L. Holzworth, M. R. Louthan, Jr. and R. G. Derrick, 1976, "The Effect of Oxygen on Hydrogen Transport in Copper", p. 370, *Corrosion*, Vol. 32.

Caskey, G. R., Jr., G. R. Cole, and W. G. Holmes, 1961, *Symposium on Powder Packed Uranium Dioxide Fuel Elements*, Windsor Conn., Nov. 30-Dec. 1, 1961, Joint U. S.-Euratom CEND-153, II, IV-E-1.

Caskey, G. R., M. R. Louthan, Jr., and R. G. Derrick, 1975, "Hydrogen Transport in Molybdenum", p. 279, *Journal of Nuclear Materials*, Vol. 55.

Caskey, G. R., and R. G. Derrick, 1974, "Trapping of Hydrogen During Permeation in FCC Metals", presented at *the American Institute of Mining, Metallurgical and Petroleum Engineers' Spring Meeting*, Pittsburgh, Pennsylvania.

Caskey, G. R., Jr., R. G. Derrick, and M. R. Louthan, Jr., 1974, p. 481, "Hydrogen Diffusion in Cobalt", *Scripta Metallurgica*, Vol. 8.

Caskey, G. R., and W. L. Pillinger, 1974, "Effect of Trapping on Hydrogen Permeation in Copper", *Hydrogen in Metals*, American Society for Metals, Metals Park, Ohio.

Caskey, G. R., and W. L. Pillinger, 1975, "Effect of Trapping on Hydrogen Permeation", p. 467, *Metallurgical Transactions*, Vol. 6A.

Costas, L. P., 1960, *Thermal Conductivity of Aluminum-Lithium Alloys Containing up to 8% Lithium*, DP-463, Savannah River Laboratory, DuPont, Aiken, SC (April 1960).

Costas, L. P., 1963, *The Diffusion of Lithium in Aluminum*, DP-813, Savannah River Laboratory, DuPont, Aiken, SC (February 1963).

Costas, L. P., and R. P. Marshall, 1962, "The Solubility of Lithium in Aluminum*", Trans Met Soc AIME*, p. 970, Vol. 224.

Donovan, J. A., 1976, "Sorption of Tritium by Nickel During Plastic Deformation", p. 145, *Metallurgical Transactions*, A, Vol. 6A.

Donovan, J. A., 1977, "Accelerated Evolution of Hydrogen from Metals During Plastic Deformation", p. 1677, *Metallurgical Transactions*, A, Vol. 7A.

Donovan, J. A., 1980, "Helium Embrittlement of Tritium Charged Metals", p. 77*, Tritium Technology in Fission, Fusion and Isotopic Applications*, U. S. Department of Energy, Document No. CONF-800427.

Hydrogen Damage, 1977, p. IX, ASM, Metals Park, Ohio.

Holzworth, M. L., and M. R. Louthan, Jr., 1968, "Hydrogen Induced Phase Transformations in Type 304L Stainless Steels", p. 110, *Corrosion*, Vol. 04.

Kanne, W. R., Jr., 1988, "Remote Reactor Repair: GTA Weld Cracking Caused by Entrapped Helium", p. 33, *Welding Journal* (August 1988).

Louthan, M. R., Jr., 1963, "Stress Orientation of Titanium Hydrides in Titanium", p. 1166, *Trans of the Metallurgical Society of AIME*, Vol. 227.

Louthan, M. R., Jr., 1974, "Effects of Hydrogen on the Mechanical Properties of Low Carbon and Austenitic Steels", p. 52, *Hydrogen in Metals*, ASM, Metals Park, Ohio.

Louthan, M. R, Jr., 1987, "Effect of Hydrogen on Metals", P. 129, *Corrosion Mechanisms*, Marcen Dekker, Inc., New York.

Louthan, M. R., Jr., G. R. Caskey, Jr., J. A. Donovan, and D. E. Rawl, 1972, "Hydrogen Embrittlement of Metals", p. 357, *Materials Science and Engineering*, Vol. 10.

Louthan, M. R., Jr., J. A. Donovan, and G. R. Caskey, Jr., 1974, "Isotope Effects on Hydrogen Transport in Nickel", p. 643, *Scripta Metallurgica*, Vol. 8.

Louthan, M. R., Jr., J. A. Donovan, and G. R. Caskey, Jr., 1975a, "Hydrogen Diffusion and Trapping in Nickel", p. 745, *Acta Metallurgica*, Vol. 23.

Louthan, M. R., Jr., J. A. Donovan, and G. R. Caskey, Jr., 1975b, "Tritium Absorption in Type 304L Stainless Steel", p. 192, *Nuclear Technology*, Vol. 26.

Louthan, M. R., Jr., G. R. Caskey, Jr., and A. H. Dexter, 1976a, "Hydrogen Effects in Aluminum Alloys", p. IV-117, *Radiation Effects and Tritium Technology for Fusion Reactors*, CONF-750098, Oak Ridge National Laboratory, Oak Ridge, Tennessee (March 1976).

Louthan, M. R., Jr., R. G. Derrick, J. A. Donovan, and G. R. Caskey, Jr., 1976b, "Hydrogen Transport in Iron and Steel", p. 337, *Effect of Hydrogen on the Behavior of Materials*, AIME, New York.

Louthan, M, R., Jr., G. R. Caskey, Jr., D. E. Rawl, Jr., and C. W. Irapp, 1976c, "Tritium Effects in Austenitic Steels", p. IV-98, *Radiation Effects and Tritium Technology for Fusion Reactors*, Vol. IV, AONF-750989, Oak Ridge National Laboratory.

Louthan, M. R., Jr., and G. R. Caskey, Jr., 1976, "Hydrogen Transport and Embrittlement in Structural Metals", p. 291, *International Journal of Hydrogen Energy*, Vol. 1.

Louthan, M. R., Jr. and R. G. Derrick, 1975, "Hydrogen Transport in Austenitic Stainless Steels", p. 565, *Corrosion Science*, Vol. 15.

Louthan, M. R., Jr., and R. P. Marshall, 1963, "Control of Hydride Orientation in Zircaloy", p. 170, *J. Nucl. Mat.*, Vol. 9.

Marshall, R. P., 1961 "Strain-Induced Diffusion and Precipitation in Aluminum-Lithium Alloy During Creep", *Acta Metallurgica*, p. 958, Vol. 9.

Marshall, R. P., 1976, "Influence of Fabrication History on Stress-Oriented Hydrides in Zircaloy Tubing" p. 34 and "Control of Hydride Orientation in Zircaloy by Fabrication Practice", p. 49, *Journal of Nuclear Materials*, Vol. 24.

Marshall, R. P., and M. R. Louthan, Jr., 1963, "Tensile Properties of Zircaloy with Oriented Hydrides", p. 693, *Trans. of the ASM*, Vol. 56.

McDonell, W. R., 1989, *Aluminum-Lithium Target Behavior*, WSRC-RP-89-970, Westinghouse Savannah River Company, Aiken, SC (October 1989).

Metals Handbook, Welding, Brazing and Soldering, 1993, Vol. 6, p. 517, ASM International, Materials Park, OH.

Morgan, M. J., 1991, "The Effects of Hydrogen Isotopes and Helium on the Flow and Fracture Properties of 21-6-9 Stainless Steel", *Morris E. Fine Symposium*, edited by P. K. Liad, J. R. Weertman, H. L. Marcus and J. S. Santner, The Minerals, Metals and Materials Society, Warrendale, Pennsylvania.

Morgan, M. J., 1997, "Tritium-Induced Cracking Model", *Proceedings 21st Aging, Compatibility and Stockpile Stewardship Conference*.

Morgan, M. J., 1999, *Tritium and Decay Helium Effects on Cracking Thresholds and Velocities in Stainless Steel*, WSRC-MS-99-00343, Westinghouse Savannah River Company, Aiken, SC.

Morgan, M. J., and D. Lohmeier, 1990, "Threshold Stress Intensities and Crack Growth Rates in Tritium-Exposed HERF Stainless Steels", p. 459, *Hydrogen Effects on Material Behavior*, edited by N. R. Moody and A. W. Thompson, The Minerals, Metals and Materials Society, Warrendale, Pennslyvania.

Morgan, M. J., and M. H. Tosten, 1990, "Microstructure and Yield Strength Effects on Hydrogen and Tritium Induced Cracking in HERF Stainless Steel", p. 447, *Hydrogen Effects on Material Behavior*, edited by N. R. Moody and A. W. Thompson, The Minerals, Metals and Materials Society, Warrendale, Pennslyvania.

Morgan, M. J., and M. H. Tosten, 1996, "Tritium and Decay Helium Effects on the Fracture Toughness Properties of Types 316L, 304L and 21Cr-6Ni-9Mn Stainless Steels", p. 873, *Hydrogen Effects in Materials*, edited by A. W. Thompson and N. R. Moody, The Minerals, Metals and Materials Society, Warrendale, Pennslyvania, 1996.

Owen, J. H., and D. Randall, 1976, "Equilibrium and Kinetic Studies of Systems of Hydrogen Isotopes, Lithium Hydrides, Aluminum and $LiAlO_2$", p. III-433, *Radiation Effects and Tritium Technology for Fusion Reactors*, CONF-750098, Oak Ridge National Laboratory, Oak Ridge, Tennessee (March 1976).

Peacock, H. B., Jr., N. C. Iyer, and M. R. Louthan, Jr., 1995, "Materials Considerations in Accelerator Targets", p. 495, *AIP Conference Proceedings 346, International Conference on Accelerator-Driven Transmutation Technologies and Applications*, edited by E. D. Arthur, A. Rodriguez, and S. O. Schriber, American Institute of Physics.

Petersen, V. C., 1982, "Hot-Salt Stress Corrosion Cracking of Titanium", p. 167, *ASM Source Book on Titanium and Titanium Alloys*, ASM, Metals Park, Ohio.

Rawl, D. E., Jr., G. R. Caskey, Jr., and J. A. Donovan, 1980, "Low Temperature Helium Embrittlement of Tritium Charged Stainless Steel", *109th Annual AIME Meeting*, Las Vegas, Nevada (February 20-23, 1980).

Rideout, S. P., R. S. Ondrejcin, M. R. Louthan, Jr., and D. E. Rawl, 1969, "The Role of Moisture and Hydrogen in Hot-Salt Cracking of Titanium Alloys", p. 650, *Fundamental Aspects of Stress Corrosion Cracking*, edited by R. W. Staehle, A. J. Forty and D. VanRooyen, NACE, Houston.

Rideout, S. P., R. S. Ondrejcin, and M. R. Louthan, Jr., 1970, "Hot-Salt Stress Corrosion Cracking of Titanium Alloys", p. 307, *The Science, Technology and Application of Titanium*, edited by R. Jaffee and N. Promisel, Pergamon Press, New York.

West, A. J., and D. E. Rawl, Jr., 1980, "Hydrogen in Stainless Steel: Isotopic Effects on Mechanical Properties", p. 69, *Tritium Technology in Fission, Fusion and Isotopic Applications*, U. S. Department of Energy, Document No. CONF-800427.

Biography

M. R. (Mac) Louthan, Jr., earned a bachelor's degree in Metallurgical Engineering from Virginia Tech in 1960 and a Ph.D. in Metallurgical Engineering and Material Science from Notre Dame in 1967.

Mr. Louthan is a senior advisory engineer in the Materials Technology Section of Savannah River Technology Center. He is the author of approximately 200 technical publications, editor of 9 books, and a Fellow in ASM International. Mac developed the lecture "Why Stuff Falls Apart", which has been given over 200 times to colleges, universities, and civic and professional organizations. He has served as president of the International Metallographic Society, a member of the Board of Trustees of the National Youth Science Foundation, and chairman or co-chairman of 12 international conferences. He was a key reader for Metallurgical Transactions, a member of the editorial advisory board for Materials Characterization, and the series editor of Microstructural Science. Mr. Louthan has given invited presentations in throughout the U.S., Canada, Europe, and Asia, and is a member of Sigma Xi, Alpha Sigma Mu and Tau Beta Pi. Mac's awards include: the President's Award and two Best Paper Awards from the International Metallographic Society; the Instructor of Merit and the Distinguished Educator Awards from ASM International; an Award of Excellence from the Federal Laboratory Consortium for Technology Transfer; the Wine and two Sporn Awards for teaching excellence at Virginia Tech; and the Orth Award and three Westinghouse Signature Awards of Excellence from the Savannah River Site.