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Hydrogen-Induced Embrittlement of Candidate Target Materials for Applications in Spallation-Neutron-Target Systems

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Project Title:
Hydrogen-Induced Embrittlement of Candidate Target Materials for Applications in Spallation-Neutron-Target Systems
(For Renewal)

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AAA Research Area: **Target / Transmuter**

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Abstract:

The purpose of this project is to evaluate the effect of hydrogen on cracking of candidate target materials for applications in spallation-neutron-target (SNT) systems such as accelerator production of tritium (APT) and accelerator transmutation of waste (ATW). The test materials will undergo appropriate thermal treatments prior to being hydrogen-charged by potentiostatic cathodic polarization technique in a simulated aqueous environment at different temperatures of interest. The specimens, upon completion of testing, will be metallographically examined. Further, the scanning electron microscopy (SEM) will be used to determine the extent and nature of cracking in the specimens tested. The thrust of the proposed testing is to determine the effect of hydrogen on environment induced cracking but not to study the effect of radiation induced damage (radiation hardening).

Work Proposed for Year 2 (Summer 2002-Spring 2003), Goals, and Expected Results:

Experimental heats of Alloys HT-9, EP-823 and 422 have been melted and processed into round bars at the Timken Research Laboratory, Canton, OH. Thermal treatments have also been completed on a batch of these materials at this vendor's facility. Test specimens are scheduled to be machined soon at a local machine shop. A large number of test equipment has been ordered using AAA-infrastructure funding, some of which have already been received from the vendors. Currently, there are two interim research laboratories at the Harry Reid Center and the Engineering Building, respectively, which will soon be consolidated into a UNLV materials research user-facility called the "Materials Performance Laboratory (MPL)" to be located in the Engineering Building. Metallurgical and corrosion studies of candidate target materials will be initiated as soon as the machined test specimens become available. It is anticipated that the first batch of testing will be performed during March 2002. Testing will be continued during the second year as outlined in the overall research plan. These tests will be followed by metallographic evaluation of tested specimens using optical microscopy and SEM. Subsequently, the resultant data will be analyzed, and reported on quarterly and semi-annual basis.

Funding Profile:

Academic Year:	2001-2002	2002-2003	2003-2004
Total (K$)	146	142	136

Background and Rationale:

During his two presentations at the AAA-UNLV Mini-Workshop held at the Harry Reid Center (HRC) on January 18, 2001, Dr. Stuart Maloy of Los Alamos National Laboratory (LANL) identified several research areas in the fields of Material Science and Corrosion Engineering. More recently, Drs. Roy and O'Toole had further interactions with Drs. Maloy and Li to follow-up on the related research topics, and to invite Drs. Maloy and Li of LANL to participate in HRC-sponsored collaborative research at UNLV in areas of Materials Science and Corrosion Engineering, respectively. Drs. Maloy and Li had expressed their willingness to collaborate with UNLV researchers and to actively participate in collaborative materials and corrosion research activities to be pursued at the Mechanical Engineering department of UNLV.

The operating environment of a SNT system can produce a considerable amount of hydrogen, helium and associated environment-induced damage in the target material. Therefore, the test specimens will be cathodically charged in a deaerated (by helium) aqueous environment of interest to evaluate their susceptibility to hydrogen-induced embrittlement (HE) at different temperatures. The test materials will be thermally treated prior to being charged with hydrogen. Thus, in essence, this project will be aimed at evaluating the effect of hydrogen content on the environment-assisted cracking of candidate target materials as a function of metallurgical conditions resulting from different thermal treatments. It is also possible to study the effect of hydrogen on the corrosion resistance of the materials. This study will provide essential information necessary to conduct future in-proton-beam radiation experiment to directly study these effects.

Test Materials:

Three martensitic stainless steels, namely HT-9, EP 823 and Type 422 will primarily be tested in this program. Ni-base austenitic Alloy 718 may also be incorporated during the testing if warranted by the AAA program needs.

Types of Testing:

The susceptibility to HE will be evaluated by using both smooth and notched uniaxial tensile specimens. The tensile specimens will be pulled in an aqueous environment under either a constant load or a slow-strain-rate (SSR) condition. For constant load testing, a loading device such as a calibrated Proof Ring will be used. For each Proof Ring, a calibration curve generated by the manufacturer will be used to determine the amount of deflection needed to apply the desired load to the test specimen. The determination of cracking tendency using this technique will be based on time-to-failure (TTF). An automatic timer attached to the test specimen will record the TTF. Tensile specimens loaded to desired stress level would provide a fail versus no fail result.

The tensile specimens will be fabricated from heat-treated plate/bar materials in such a way that the gage section is parallel to the longitudinal rolling direction. For constant-load testing, the magnitude of the applied stress will be based on the tensile yield strength (YS) of the test material. The test specimens will be loaded at different percents of YS and the corresponding TTF will be recorded.

During SSR testing, the specimen will be continuously strained in tension until fracture, in contrast to more conventional SCC testing conducted under a sustained load condition. The application of slow dynamic straining (10^{-6} to 10^{-7} sec^{-1}) to the smooth tensile specimen will facilitate cracking in the test materials that may not exhibit cracking under constant load or may take prohibitively long duration for crack initiation. However, the use of a notched tensile specimen may result in fast crack initiation under both constant load and SSR conditions. The smooth tensile specimen in SSR testing may undergo fast failure in a ductile manner if SCC does not occur, or prematurely in a brittle manner if SCC occurs. Prior to being tested in the aqueous environment, a few SSR specimens (both smooth and notched) will be pulled in air to establish baseline data.

It is well known that electrochemistry can play an important role in characterizing the stress corrosion cracking (SCC)/hydrogen embrittlement (HE) behavior of susceptible alloys. Accordingly, at the onset, the corrosion or the open circuit potential (E_{corr}) of the test specimen will be measured in the aqueous environment of interest with respect to a standard Ag/AgCl reference electrode. The specimen will then be strained in the test solution either in a constant load or in a SSR condition using either a Proof Ring or a mechanical testing machine (Instron or MTS) with or without a controlled electrochemical potential (E_{cont}). The magnitude of E_{cont} will be selected based on the measured E_{corr} value (cathodic versus E_{corr}). This cathodic (negative) potential (E_{cont}) will be applied potentiostatically during the entire straining period. Two graphite rods will be used as cathodes during the potentiostatic polarization of the tensile specimen (anode). A three-electrode polarization technique will be used.

For conventional SCC tests under static loading condition, the cracking susceptibility will be expressed in terms of a threshold stress (σ_{th}) below which cracking does not occur during arbitrary test duration, or the TTF at a given applied stress. With SSR tests, however, similar approaches cannot be used since the test specimen is subjected to a continuously changing stress during straining. Therefore, the cracking tendency will be characterized by a number of readily measurable and quantifiable parameters obtained from the load-deflection curve and the examination of the broken test specimen.

Since environment-assisted cracking is usually associated with relatively little macroscopic plastic deformation during crack growth, the ductility parameters such as the percent elongation (%El) and the percent reduction in area (%RA) are very useful in characterizing the cracking susceptibility. In

addition, the true fracture stress (σ_f) obtained from the load-deflection curve and the final specimen dimension is useful in expressing the extent of cracking tendency.

Secondary ion mass spectrometry (SIMS) will be used to analyze the hydrogen content resulting from the cathodic potentiostatic polarization. Optical microscopy and scanning electron microscopy (SEM) will be used to characterize the extent and morphology of cracking in all materials tested. Energy dispersive spectroscopy (EDS) will be used to detect and identify chemical species present in the vicinity of cracks.

Test Conditions:

All three martensitic stainless steels will be received in the form of both bar and plate. This group of materials is called martensitic because they are capable of changing their crystal structure upon heating and cooling. They can be quench hardened to fully martensitic structures without any retained austenite. These plates and bars will be austenitized in the temperature range of 1900 to 2000^0F for 1 hour followed by a water/oil quench. These martensitic structures are very hard and brittle. Therefore, these quenched materials will further be tempered at 1200^0F for 1, 1.5 and 2 hour(s) followed by air-cooling. All test specimens will be fabricated from these quenched and tempered stainless steels.

The susceptibility to HE will be evaluated by subjecting the thermally treated tensile specimens to cathodic charging in a deaerated (by helium) acidic brine (NaCl solution) at temperatures ranging from ambient (25^0C) to elevated temperatures (up to 120^0C). The strain rate used during the SSR tests will range from 10^{-6} to 10^{-7} sec^{-1}. The pH of the test solution will range between 2 and 3. Duplicate specimens of each material will be tested under each heat-treat and electro-chemical conditions.

Expected Data:

The proposed experimental program will generate the following data:

- Corrosion potential (E_{corr})
- Failure load (P_f)**
- True fracture stress (σ_f)**
- Threshold stress for cracking (σ_{th})**
- Percent elongation (%El)**
- Percent reduction in area (%RA)**
- Time-to-failure (TTF)**
- Hydrogen content evaluated by SIMS
- Metallurgical microstructures evaluated by optical microscopy
- Fracture Characteristics evaluated by scanning electron microscopy

**Versus E_{cont}

The resultant data will enable a mechanistic understanding of the HE process in candidate target materials under the synergistic influence of applied tensile stress and the corrosive media with or without the presence of hydrogen. Others may eventually utilize these data in developing and validating the hydrogen-induced degradation models for target materials used in transmutation processes.

Research Capabilities at UNLV

The UNLV AAA Infrastructure Committee has reviewed the need for some state-of-the-art equipment to accomplish the proposed metallurgical and corrosion research goals. The following equipment has recently been ordered using funds approved by this committee, some of which have already arrived at the campus and will soon be installed at MPL.

- Multi-channel potentiostat/Corrosion software for electrochemical studies (PerkinElmer Inc.)
- Proof ring assemblies for constant-load SCC testing (Cortest Inc.)
- Constant-extension-rate-testing (CERT) system for SSR testing (Cortest Inc.)
- Electrochemical cells/water bath/electrodes etc.
- High-resolution (1000X) inverted optical microscope for metallographic evaluation (Leica)
- Abrasive cutter for sample preparation (Buehler)
- Heat-treating furnace (1200^0C) with larger working dimensions (Lindberg)
- High-temperature inert gas chamber for mechanical testing (MTS)
- Other laboratory-related parts and equipment

Heat Treatment Facilities
Two high temperature furnaces are available:

1) Lindberg Furnace
The maximum temperature is 1200 ^{0}C (2200 ^{0}F). The working dimensions are 15” x 7.5” x 5.5”.

2) Thermodyne Furnace
The maximum temperature is 1200 ^{0}C (2200 ^{0}F). The working dimensions are 6.5” x 4.5” x 4.5”.

Machine Shop
The UNLV College of Engineering has a machine shop with two vertical mills, two lathes, a welding station, and a variety of band saws, shear breaks, and drill presses. None of this equipment is automated so we have developed good working relationships with several local machine shops. There are several good local shops with CNC, EDM, water jet, and laser cutting capabilities that can be contracted at reasonable rates.

Microstructural Analysis
The UNLV Mechanical Engineering Department has a photomicroscopy lab with two 3-wheel sample polishing stations along with a sample potting machine and sanding wheels. The lab has a Unimet Unitron 8644 Inverted Metallurgical Microscope with 800X magnification equipped with a digital camera and computer for recording micrographs. The lab also has a Leco M-400A microhardness tester, several Wilson and Clark Rockwell hardness testers, and a Beuler sample mounting press.

However, the metallographic laboratory needs to be upgraded with a high-resolution optical microscope and accessories to perform microstructural characterization planned in this project. Analyses of hydrogen content by SIMS can be performed at LANL.

Scanning Electron Microscopy (http://www.unlv.edu/Colleges/Sciences/Geoscience/EMIL.htm)
The UNLV Geosciences Department has a JEOL-5600 Scanning Electron Microscope (SEM). It is optimized for imaging micron to millimeter scale topographic detail of solid materials. Resolution of up to 50 nm at 100,000 times magnification is possible. The SEM is equipped with a BSE detector and

an Oxford ISIS EDS system, capable of semi-quantitative analysis (± 10%). The topographic and compositional images can be processed directly on the screen to show pseudo-color and critical point measurement of features. The images can also be combined, allowing for easy comparison of samples or different magnifications. The manual stage can accommodate four 1-cm diameter samples or one sample up to 3.2-cm diameter. The SEM and EDS are controlled by two networked Windows 95 operating systems allowing for intuitive, simple operation.

The UNLV Geosciences Department also has the JEOL-8900 Electron Probe Microanalyzer (EPMA). It is optimized for quantitative, non-destructive chemical analysis of solid materials on a micron scale. Four fully automated wavelength dispersive spectrometers (WDS) are equipped with 2 crystals each and are capable of quantifying elements ranging from boron to uranium. Concentrations of at least 0.10 wt % can be measured to within ±1% of the measured abundance. In addition, elements present in smaller concentrations can be measured with somewhat less precision. The energy dispersive spectrometer (EDS) collects a full spectrum of x-rays at once and is capable of rapidly qualifying up to 8 elements at one time. Both EDS and WDS can also be used to obtain high-precision x-ray maps and line scans of spatial variation in chemical composition. The instrument is also equipped with backscattered electron, secondary electron, and cathodoluminescence detectors capable of producing "real time" images, or automated images in tandem with x-ray mapping to further characterize the area of interest. A fully automated stage, capable of holding up to nine one-inch round samples (or six petrographic sections) has reproducibility of less than one micron. Unmounted samples up to 15 cm in diameter can also be accommodated. The EPMA is controlled by a graphical user interface on a HP-UX UNIX workstation. These two instruments are available as a user facility. A fee structure has currently being developed.

Transmission Electron Microscopy

A transmission electron microscopy (TEM) is planned to be established as part of the UNLV AAA program. The UNLV AAA Infrastructure committee is currently evaluating the capabilities of three different TEM manufacturers, and a procurement order will be placed with the successful bidder. The anticipated date for the establishment of this facility is in the fall of 2002.

Project Timeline

Timeline Narrative

This research project started during the summer of 2001. Mr. Raymond Kozak, who has been working in this project as a Ph.D. level graduate research assistant since its inception, recently decided to discontinue his graduate study due to medical and personal reasons. Efforts are ongoing at this time to replace Mr. Kozak with another competent graduate student to accomplish the planned research goals. It is anticipated that the new replacement graduate student will be identified by May 2002. Meanwhile, Mr. Konstantin Zabotkin, a recent UNLV M.S. graduate in mechanical engineering, has been added to this program to expedite the planned experimental program. Mr. Mark Jones, who is pursuing his Master’s degree in mechanical engineering, is still continuing in this project to complete his M.S. thesis on this project topic. Additionally, Mr. Aaron Tippetts, an undergraduate student in mechanical engineering, has recently joined this project to assist in experimental work and other project-related tasks. The UNLV research team (faculty and students) will visit LANL during 2002 spring to meet the collaborator(s) from this national laboratory to refine our testing plan, and be familiar with related projects at LANL. This visit will enhance coordination and cooperation among researchers from both UNLV and LANL.

Current efforts are being focused on machining different types of test specimens using heat-treated bars. Machining of the test specimens will be done at a local shop having precision machining capabilities. Simultaneously, the final testing laboratory is being organized to accommodate both the existing and future equipment so that all desired experimental activities could be performed successfully in a timely fashion in the most cost-effective manner. The tested samples will be evaluated by optical microscopy and SEM. The effect of hydrogen content on the cracking susceptibility will be evaluated. Attempts will be made to correlate the resultant cracking parameters to the microstructures of the different stainless steels under different metallurgical conditions. It is anticipated that a substantial part of testing will be completed by the end of the second year. Reports will be prepared on quarterly and semi-annual basis. A two-year schedule is shown in Table 1.

Table 1: Two-Year Research Plan

Time (Months)	3	6	9	12	15	18	21	24	27	30	33	36
Literature Search	■	■										
Order Materials	■											
Order Equipment	■											
Set-up Laboratory	▌	■	■	■								
Heat Treat Materials		■	■									
Machine Samples		▌	■	■	■							
Train Students	■	■	■	■	■							
Perform Experiments				■	■	■	■	■	■			
Perform Microscopy					■	■	■	■	■	■	■	
Perform SEM					■	■	■	■	■	■	■	
Data Analyses					■	■	■	■	■	■	■	▌
Follow-up Proposal												■
Quarterly Reports	■	■	■	■	■	■	■	■	■	■	■	■
Semi-Annual Reports		■		■		■		■		■		■

Year 2 Milestones (Assuming a start date of May 20, 2002)

- (May 2002) Construction of the Materials Performance Laboratory has been completed.
- (June 2002) Test specimens have been machined from heat-treated bars
- (May 2002) SCC testing has been initiated without hydrogen charging using both proof ring and SSR techniques.
- (July 2002) HE testing (cathodic charging) has started.
- (August 2002) Optical microscopy and SEM evaluation have started. Data analyses are initiated.
- (May 2003) Year 2 report is submitted.

Deliverables

- **Collaboration with DOE project:** Monthly communications (by phone or in person) with National Project collaborator and/or technical lead to update on progress, discuss problems, and allow for re-focusing if necessary to address shifts in direction by the National Project.
- **M.S. Degree:** Graduate students will have completed or be very close to completion of their experiments and analyses by the end of the second year.
- **Progress Reports:** Brief reports indicating progress will be provided every quarter (to support DOE AAA quarterly meetings).

- **Bi-Annual Reports:** Written reports detailing experiments performed, data collected and results to date.
- **Final Report:** Written report detailing experiments performed, data collected, results, and conclusions to be submitted at the end of the project.
- **Project Samples:** For archival purposes, samples generated during the experimental campaigns will be turned over to the National Laboratory partner. For experiments where multiple samples were prepared, only one sample will be turned over. This sample archive will allow the Project researchers (either from the National Laboratories or UNLV or other academic partners) to re-examine samples as necessary, either in support of this work or for use in other research projects.

Personnel Responsibilities

Role of the Students:

The graduate students will be given a significant amount of responsibility for this project. Mark Jones (M.S. student) has a significant amount of experience machining test samples, polishing samples for microscopy, and conducting mechanical testing experiments. However, both Mark and the other new graduate student (to be hired) will undergo training in areas of basic metallurgical and electrochemical concepts including heat-treating of metallic materials, electrochemical polarization, preparation of SCC/HE tests, and metallographic evaluation using optical microscopy and SEM. The faculty investigators will provide these training. The new graduate student will be advised to take relevant metallurgical, mechanical, and corrosion engineering basic and advanced level courses. Both students will prepare and present their research objectives by August 2002.

An undergraduate research assistant will also be hired to help with this project. It is important to involve undergraduates with research so that they are exposed to research opportunities. This student will help with heat treatments, machining, sample preparation testing, and microstructural analysis, as needed.

Role of the Faculty:

The primary responsibility of the faculty members on this project is to train students for performing the various research tasks, supervise the writing of their thesis and/or dissertation, and to provide an interdisciplinary working environment for the students. Dr. Roy joined the Mechanical Engineering Department in July 2001 as an associate research professor, concentrating on both teaching and funded research projects. Fifty percent (50%) of his compensation is funded by the AAA program. The faculty members will assume responsibility for various aspects of the project to ensure that the students make steady progress towards their research goals. The faculty will not be doing the research for the students but will ensure that high quality work is accomplished in a timely manner. Each of the faculty advisors has unique expertise in one or more fields related to this project. The faculty will also be learning from each other.

As the lead principal investigator (PI), Dr. Roy will spend 25% of his time in this project. He will work in close association with Dr. O'Toole in areas of mechanical aspects related to the SSR testing unit. Dr. Roy will be responsible for specifying and ordering test materials from prospective vendors. The students will be included in this process from the beginning. Drs. O'Toole and Roy will perform ordering of the test equipment and setting-up of the laboratory jointly. During the initial phase of this project, Dr. Roy will train the graduate students in areas of electrochemical test techniques, heat-

treating practices, microstructural and fractographic analyses. Later, Drs O'Toole and Roy will train the graduate students on the practical aspects of performing the HE experiments using the SSR method. Dr. Roy will take the lead in specifying and ordering the test specimens from an outside vendor. Dr. Roy will work closely with the graduate students and other supporting staff within the UNLV infrastructure to ensure that the desired data are generated in a timely manner. The graduate students under the direct supervision of both Drs. O'Toole and Roy will record all test data in the scientific notebooks. Drs. Roy and O'Toole will jointly perform data analyses and prepare the interim and final reports. Dr. Hatchet will be responsible for the electrochemical cells and microstructural analysis. Dr. Wang will be responsible for the microstructural and fracture mechanics analysis.

Date: February 12, 2002
From: Ning Li, Ph.D., Los Alamos National Laboratory

To Whom It May Concern:

Since the commencement of the project **"Hydrogen-Induced Embrittlement of Candidate Target Materials for Applications in Spallation-Neutron-Target Systems"** in 2001, I have met and discussed the progress and status with Drs. Roy and O'Toole several times during my visits to UNLV and in phone conversations. The focus was mostly on material and equipment procurement, and facility setup in preparation of the test experiments.

The project has received the materials, and equipped a lab with the necessary test instruments, and is ready to perform the main task of testing. The year 2 renewal proposal contains the most important tasks for this project. I strongly recommend the renewal of the project, and look forward to receiving the reports on experimental findings.

Yours truly,

Ning Li