ANNUAL RESEARCH REPORT

2009

FOR RESEARCH PROGRAMS CONDUCTED IN 2008

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The Annual Research Report

This report summarizes the principal research activities in the Michigan Ion Beam Laboratory during the past calendar year. Fifty-six researchers conducted thirty-one projects at MIBL that accounted for over 3200 hours of equipment usage. The programs included participation from researchers at the University, corporate research laboratories, private companies, government laboratories, and other universities across the United States. The extent of participation of the laboratory in these programs ranged from routine surface analysis to ion assisted film formation. Experiments included Rutherford backscattering spectrometry, elastic recoil spectroscopy, nuclear reaction analysis, direct ion implantation, ion beam mixing, ion beam assisted deposition, and radiation damage by proton bombardment. The following pages contain a synopsis of the research conducted in the Michigan Ion Beam Laboratory during the 2008 calendar year.

About the Laboratory

The Michigan Ion Beam Laboratory for Surface Modification and Analysis was completed in October of 1986. The laboratory was established for the purpose of advancing our understanding of ion-solid interactions by providing up-to-date equipment with unique and extensive facilities to support research at the cutting edge of science. Researchers from the University of Michigan as well as industry and other universities are encouraged to participate in this effort.

The lab houses a 1.7 MV tandem ion accelerator, a 400 kV ion implanter, and an ion beam assisted deposition (IBAD) system. Additional facilities include a vacuum annealing furnace, a surface profilometry system, and a scanning laser surface curvature measurement system. The control of the parameters and the operation of these systems are mostly done by computers. They are interconnected through a local area network and the world wide web, allowing off-site monitoring and control.

Respectfully submitted,

Gary S. Was
Director
IRRADIATION EFFECTS ON STRESS CORROSION CRACKING OF AUSTENITIC FUEL CLADDING

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Spent AGR nuclear fuel bundles are stored in ponds prior to reprocessing. These may be susceptible to intergranular stress corrosion cracking (IGSCC). Mitigation is through the addition of sodium hydroxide as a corrosion inhibitor. IGSCC is attributed to chloride-induced corrosion at chromium depleted grain boundaries. Chromium depletion is a result of Radiation Induced Segregation (RIS). This project is aimed at simulating RIS levels in 20/25 Nb stabilized stainless steel cladding material by thermal sensitization and proton irradiation and exploring corrosion behavior in simulated pond water environments.

Cladding material was proton irradiated using an irradiation energy of 2.2 MeV and a dose rate of $10^{-5}$ dpa/s to a penetration depth of 23μm. Irradiations to 3 and 5 dpa at 400°C were performed.

Analytical transmission electron microscopy has been used to examine grain boundary compositions in proton irradiated as well as thermally sensitized material using Energy Dispersive X-ray (EDX) Analysis. Proton irradiated samples have shown lower and more localized levels of Cr depletion when compared with thermally sensitized material (Figure a). Proton irradiated material also showed evidence of matrix damage (Figure b).

(a) Comparison of typical chromium profiles across grain boundaries in thermally sensitized (HT2) and proton irradiated material to 3 and 5dpa. (b) Transmission electron micrograph of proton irradiated material (3dpa).
HOST-ION INTERACTIONS IN ERBIUM-DOPED WAVEGUIDE AMPLIFIERS: REGARDING THE USE OF NON-EQUILIBRIUM PROCESSING TECHNIQUES

G. Bernard and J. Kieffer, University of Michigan

Active optical waveguides, such as erbium-doped waveguide amplifiers (EDWAs), rely on adequate spatial dispersion of the active ions within the host material to inhibit non-radiative decay and, therefore, provide maximum lasing efficiency. The low solubility of erbium in silica glass and its tendency to agglomerate continue to be problematic for the production of these devices. Our hypothesis was that non-equilibrium processing techniques, namely sol-gel synthesis and ion implantation, would be able to overcome these problems and allow us to achieve higher erbium concentrations.

In this study, erbium ions were implanted at a dose of $4.4 \times 10^{15}$ ions/cm$^2$ in order to achieve a peak concentration of $\approx 1$ at. % in both fully dense fused quartz ($\rho=2.32$ g/cc) and highly porous silica aerogel material ($\rho=0.17$ g/cc). Aerogel, which is formed through the super-critical drying of sol-gel derived glass, exhibits a large pore volume and, based on the fractal nature of the silica network, a wide pore size distribution. This in turn results in a wide distribution of implantation stopping lengths, and thus better dispersion of the implanted erbium. Our process could even lead to Er$^{3+}$ concentrations exceeding the thermodynamic solubility limit in silica.

Implanted samples were investigated using Rutherford Backscattering Spectroscopy ($\text{He}^{++}$, 2 MeV, and backscattering angle of 165°) and the results are shown in the figure below. As is apparent, the erbium implanted into the porous aerogel has a much wider distribution and lower peak concentration than in fused quartz, confirming the fundamental proposition of our experiment. Because the distributions shown below are relative to the density of the respective materials, it is expected that the sintering of implanted aerogel for use in active optical waveguides will not affect the improved spatial distribution of erbium ions, leading to devices with enhanced lasing efficiency. Experiments to confirm this are planned for the future.

![RBS plots for erbium ion-implanted fused quartz and silica aerogel material. Fit lines were created using SIMNRA program.](image)

The work has been supported in part through a Rackham Merit Fellowship from the University of Michigan and in part through a grant from the National Science Foundation No. NSF-DMR 0605905.
EFFECTS OF N-BASED POINT DEFECTS ON THE ELECTRONIC PROPERTIES OF GaAsN

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Dilute nitride semiconductor alloys are promising for a wide range of applications, including long-wavelength light-emitters and detectors, high performance electronic devices, and high efficiency solar cells. For GaAsN and InGaAsN alloys, the electron mobility and optical emission efficiency decrease as the N incorporation increases, presumably due to charge trapping, scattering, and/or recombination at N-based point defects. Understanding and control of these defects would provide an opportunity to increase the minority carrier lifetime, thereby improving the performance of solar cells and transistors. Therefore, we are examining the influence of the N interstitial and the N\textsubscript{As}-Si\textsubscript{Ga} complex on the electronic properties of GaAsN alloys.

It has been reported that GaAsN alloys contain a significant fraction of interstitial N,\textsuperscript{1} shown in (a). A few studies have shown that annealing reduces the interstitial N fraction, presumably due to N diffusion to As vacancies. However, the influence of interstitial N on the electronic properties of GaAsN alloys remains unknown. Thus, we are using annealing to probe the influence of N interstitials on the electronic properties of GaAsN. In as-grown GaAsN films, temperature-dependent Hall measurements reveal a thermally activated increase in [n] for temperatures > 150 K, suggesting the presence of a defect level below the conduction band edge, presumably due to interstitial N. Upon annealing, [n] increases and becomes nearly temperature-independent, indicating a decrease in the concentration of trapping centers, presumably due to a reduced concentration of interstitial N.

In GaAs-based materials and devices, silicon is the most common n-type dopant. However, in dilute nitride alloys, it has been suggested that Si and N atoms form N\textsubscript{As}-Si\textsubscript{Ga} complexes, shown in (b). Here, we report the first quantitative evidence of N-Si complex formation by comparing the properties of GaAsN films doped with Si and Te, with a variety of N-dopant spatial separations. First, we compare bulk-like GaAsN:Si films, where Si and N reside within the same layer, with modulation doped heterostructures, where N and Si atoms are separated spatially. A decrease in free carrier concentration, [n], with increasing N composition is observed in bulk-like films but not in the heterostructures, suggesting N-Si defect complexes in bulk GaAsN layers are likely acting as trapping centers. In addition, we compare GaAsN films doped with Si and Te. For GaAsN:Te films, [n] increases substantially with increasing annealing temperature, but little change is observed in GaAsN:Si films, as shown in (c). In GaAsN:Si, the annealing-induced increase in [n] is presumably balanced by the formation of additional Si\textsubscript{Ga}-N\textsubscript{As} complexes.

This project was supported by the National Science Foundation NSF-FRG grant, No. DMR-0606406.
An optical isolator is a component that transmits light efficiently in one direction but prevents backward propagation. Integration of optical isolators with other optical devices is desired because they can improve on-chip optical systems and reduce device size and cost. On-chip optical interconnect technology requires a high data rate with a large signal-to-noise ratio, so unwanted backward propagation should be minimized.

Research on optical isolators has been extensive in the past decade, leading to a wide range of isolator designs. The most traditional design consists of a Faraday rotator and two polarizers, but the polarizers makes these devices complicated for on-chip integration. To avoid this drawback, in the past few years a new generation of isolators have been developed that operate on the magneto-optical phenomenon of a nonreciprocal phase shift, rather than polarization conversion-based Faraday rotators. Such designs include Mach-Zehnder interferometers (MZIs) and single waveguide dielectric or photonic crystal (PC) isolators. We propose a novel isolator design that both: (1) operates via backward propagating mode cutoff and (2) employs a simple dielectric waveguide design. Furthermore, we achieve single mode, unidirectional propagation in plane wave expansion (PWE) simulations. Our design offers several advantages over previous designs: MZIs require precise interference that is difficult to achieve, PCs are larger structures and the dielectric waveguides in the literature are both multimode and operate on nonreciprocal absorption loss, which requires active isolator components. In order to prove this idea, we use the Bismuth ion implantation, provided by the Michigan Ion Beam Laboratory, to high-index optical waveguide (figure below). The implant conditions were: Bi$^+$ ions, energy 400 KV, room temperature and fluences between $5 \times 10^{14}$ ions/cm$^2$ and $2.5 \times 10^{15}$ ions/cm$^2$. We are currently evaluating the optical isolators.
AstroWatt is developing layer transfer technologies for enabling solar cell formation on flexible substrates. Using this approach, device layers are implanted with high energy ions capable of blistering layers within a semiconductor substrate. Using wafer bonding and layer transfer approaches, solar cells are formed on flexible substrates. Implantation of various ion species at different energies using the 400 kV NEC ion implanter was performed to enable development of this technology.
IRRADIATION ASSISTED STRESS CORROSION CRACKING OF AUSTENITIC ALLOYS IN SUPERCRITICAL WATER

R. Zhou, E. A. West, Z. Jiao, and G. S. Was
University of Michigan

The supercritical water reactor is one of the promising Generation IV nuclear reactor concepts. One of the greatest challenges for SCWR, however, is identifying materials for fuel assemblies and internal components in the aggressive SCWR environment. The irradiation assisted stress corrosion cracking susceptibility of four commercial austenitic alloys D9, 316L, 690, and 800H was evaluated in 500°C deaerated supercritical water. The alloys were irradiated with 2 MeV protons to a dose of 7 dpa at a temperature of 500°C. All four alloys exhibited irradiation induced hardening. Alloy 316L showed the highest hardening induced by irradiation and D9 showed the lowest. Constant extension rate tensile experiments were performed on the irradiated specimens in 500°C deaerated supercritical water. The results showed that all four alloys were susceptible to irradiation assisted stress corrosion cracking. Alloy 690 behaved relatively well in the unirradiated condition, but underwent a dramatic increase in cracking in the irradiated condition. The cold-worked D9 stainless steel displayed the least amount of intergranular cracking. The average crack length was 19, 39, 55 and 145 μm for D9, 316L, 690, and 800H respectively. It can be seen from Figure 1 that the average crack length of alloys 690 and 800H is larger than that of alloys 316L and D9. Figure 2 shows that the crack length per unit area was 425, 1905, 8426, and 2070 for alloys D9, 316L, 690, and 800 H respectively.

This Project is supported by the U.S. Department of Energy under NERI grant DE-FC07-05ID14664.
LOCALIZED DEFORMATION AS A PRIMARY CAUSE OF IRRADIATION ASSISTED STRESS CORROSION CRACKING

Z. Jiao and G. S. Was, University of Michigan

Localized deformation is a potential factor in irradiation assisted stress corrosion cracking (IASCC) of austenitic stainless steels in LWR environments. To gain a better understanding of the mechanism of IASCC, Seven austenitic alloys with a wide spread in stacking fault energy (15 – 61 mJ/m²) were irradiated with 2–3 MeV protons at 360°C to doses of 1 to 5 dpa. Tensile samples were strained in simulated BWR water at a constant extension rate of 3.5×10⁻⁷/s, and were interrupted at 1% and 3% strain. The degree of cracking was characterized using scanning electron microscopy (SEM). Parallel tensile experiments were also carried out in an argon atmosphere and the degree of localized deformation was determined by measuring the height of dislocation channels emerging on the surface using atomic force microscopy. Localized deformation contributes to IASCC by promoting grain boundary deformation at the intersections of large dislocation channels and grain boundaries. The results show that IGSCC susceptibility of austenitic stainless steels is dependent upon the degree of localized deformation.

Support for this research was provided by the U.S. Department of Energy under grant DE-FG07-05ID14703 and EPRI through the Cooperative IASCC Research (CIR) program.

Effect of localized deformation as measured by the weighted average channel height on the propensity for IGSCC as measured by crack length per unit area and strain.
IMPROVING THE SENSITIVITY OF A BIOSENSOR BY OPTIMIZING DEPOSITED SILICON THICKNESSES

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We have developed a novel optical sensor for biomedical application. It is based on multiple dielectric layers and designed to have a structure of A(BA)³X, where A, B represent high-index and low-index materials, separately. The X material is used as the defect layer and should have suitable absorption so that there will be a resonance dip in the reflectance spectrum (Figure 1). If the band width of the resonant dip is narrow and the minimum reflectance is small, then the detection sensitivity of the biosensor is high.

The initial work was done at MIBL using the Ion Beam Assisted Deposition system (IBAD) by depositing alternate layers of SiO₂, TiO₂ and Si on a silica substrate. The structure of the filters is composed of 106 nm-TiO₂ and 334 nm-SiO₂ as pairs of alternating dielectric layers. Pure silicon is used as absorptive material of the defect layer and silica as the rest of the defect layer. We tested a series of filters with different silicon thickness (from 2 to 50 nm) with suitable silica thickness (tuning resonant dip to our interested wavelength 632.8 nm), and found out that 18 nm Si thickness and 320 nm-SiO₂ produced an optimized resonant dip with Rmin = 0.43 & Δλ = 1.20 nm, which is in agreement with the simulation using a transfer matrix treatment (Figures 1 and 2 below).

This project has been funded with Federal funds from the National Institutes of Health under RR021893. GYB acknowledges the support of the Riethmiller Fellowship.
Carbon Nanotubes (CNTs) are increasingly becoming a vital material for various applications ranging from biomedical to aerospace technology. Atmospheric pressure chemical vapor deposition (CVD) is used for growing vertically aligned CNTs off the surface of a multi-layered thin film catalyst composed of 1 nm Fe deposited on top of a 10 nm Al₂O₃ buffer layer. Recycling of these catalysts was previously carried out; nevertheless, the throughput of CNTs grown on recycled catalysts proved to be much lower than freshly deposited ones. Thus, the diffusion model shown in the figure was proposed to explain this decay of catalyst activity with repeated growth.

Rutherford Backscattering (RBS) performed at MIBL was used to test the hypothesis of Fe particle diffusion into the layers below it. A helium ion beam of 2-MeV energy was aimed normal to the samples with an inclination angle of 165° between the beam and the detector. Initial results are shown in the spectrum in the figure.

RBS spectrum for 1 nm Fe / 10 nm Al₂O₃ on SiO₂ coated Si wafer.
FLUENCE ACCURACY AND UNIFORMITY USING THE 400-KV IMPLANTER AT MIBL

F. Naab and O. Toader
Michigan Ion Beam Laboratory, University of Michigan

Two different implants were performed with the 400-kV ion implanter at MIBL in order to assess the accuracy and uniformity of the fluence delivered to the sample using the Rutherford Backscattering Spectrometry (RBS) technique.

400-keV Cr\(^{+}\) ions were implanted in a Si wafer sample. The fluence delivered to the sample was calculated to be \(2.47 \times 10^{16}\) Cr/cm\(^2\) using the integrated charge from the 4-point Faraday cups and their known areas. Then, the fluence was measured using RBS with 2-MeV He\(^{++}\) ions using the 30° beamline of the 1.7 MV tandem accelerator at MIBL. The value measured using RBS was \(2.34 \times 10^{16}\) Cr/cm\(^2\). Typical uncertainties in RBS measurements are ±10%. The difference between the two measured values is 5.2% which is within the uncertainty interval. These measurements demonstrate that there is very good control of the fluence delivered to the sample using the 4-point Faraday cup system.

A second implant was done in a 2''-diameter Si wafer using Fe\(^{+}\) ions at 100 keV to \(1.0 \times 10^{16}\) Fe/cm\(^2\). After the implant the sample was subject to RBS analysis under the same conditions mentioned above. The plot below shows a schematic of the 2''-diameter Si wafer with the different spots that have been tested and labeled from 1 to 12. The Fe RBS signal from each spot was divided by the beam integrated charge through the target. Then the average value was calculated and used to normalize them. The plot below shows the normalized fluence for each spot. The error bars are the statistical uncertainties of the Fe RBS signal. The results show a very uniform implant.

![Normalized Fe\(^{+}\) fluence in the 2 inch Si wafer.](image-url)
DEVELOPMENT OF MANAGEMENT TECHNOLOGY FOR IASCC IN REACTOR INTERNALS

Korea Atomic Energy Research Institute, Korea

There are 20 nuclear power plants in Korea, which generate about 40 percent of its total electricity demand. They are 16 pressurized water reactors and 4 pressurized heavy water reactors.

The oldest unit is Kori-1 which had its license renewed in 2007 after a 30 year operation. As degradation of reactor internals during an operation has been reported, and radiation induced degradation of the stainless steel parts in these reactor internals might be an issue if such plants are power-uprated for better efficiency; such degradation needs to be evaluated carefully. In addition, international cooperation research programs have been undertaken to resolve the radiation induced degradation, IASCC (Irradiation Assisted Stress Corrosion Cracking), swelling, and hardening.

Thus, research is needed to establish the management technology for IASCC in reactor internals in Korea. Therefore a government funded IASCC research project began in 2007 and this project covers the following:

- Calculation of the neutron fluence for candidate reactor internals
- Data analysis on the IASCC history of stainless steels
- Development of an IASCC test facility
- Development of a technique for radiation-simulated test specimens
- Evaluation of an ion implantation / He irradiation effect on Stainless steels
- IASCC test in a simulated PWR water environment

We irradiated stainless steel samples with 2 MV protons to doses of 1, 3 and 5 dpa at 360°C. Currently we are conducting the analysis of the irradiated samples.
The purpose of this work is to measure the thickness of ZnO thin film grown on Si substrates, and to establish a relationship between the thickness and the deposition conditions such as deposition time. It is an essential step for further measurement of electrical properties of the films.

ZnO films were deposited by pulsed laser deposition onto Si substrates. The film thickness was measured using a Dektak II step profiler at Michigan Ion Beam Laboratory (MIBL). Each sample was measured three to four times and the average value was taken as the film thickness.

The table shows the average thickness of ZnO films deposited under different growth time. All the thickness data are used as references in electrical measurements.

Average thickness of ZnO films (nm).

<table>
<thead>
<tr>
<th>Dep. Time (mins)</th>
<th>Film thickness (nm)</th>
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<tr>
<td>60</td>
<td>650</td>
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<td>40</td>
<td>500</td>
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<td>30</td>
<td>260</td>
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ACCELERATOR BASED STUDY OF IRRADIATION CREEP IN PYROLYTIC CARBON USED IN TRISO FUEL PARTICLES FOR THE VHTR

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This work has focused on performing initial proton irradiation induced creep experiments in pyrolytic carbon (PyC). Pyrolytic carbon is the primary coating material for the TRISO fuel particles for the Gen-IV Very High Temperature Reactor (VHTR). Fission gas buildup within the TRISO particles induces a stress on the inner PyC layer, which when combined with the high operating temperature and neutron irradiation damage, can result in irradiation creep.

The first irradiation induced creep experiment was recently performed, which showed that the irradiation stage is working as intended, which includes the in situ measurement of sample elongation using the laser speckle extensometer and the monitoring of sample temperature with the infrared thermal imager. The figure below shows the creep results for the experiment, which was performed with 2MeV protons, at 900ºC, with an initial applied stress of 36MPa, and a beam current density of 5μA/cm². The expected creep rate was on the order of 10⁻⁷s⁻¹, and the average measured creep rate was 1.6x10⁻⁷s⁻¹.

This work is supported by the Department of Energy under NERI grant DE-FC07-06ID14732.

Creep strain on PyC irradiated with 2MeV protons at 900ºC with a 36MPa stress resulting in an average creep rate of 1.6x10⁻⁷s⁻¹ (red line), average irradiated sample temperature (blue line), and beam current density (green line).
The supercritical water reactor (SCWR) is a promising Generation IV nuclear reactor concept. Its benefits of increased efficiency and simplified plant design, however, come at the expense of a severe reactor environment. It is anticipated that core components will be exposed to temperatures as high as 620°C and doses as high as 30 dpa. The combination of temperature, stress, and irradiation damage creates the potential for irradiation assisted stress corrosion cracking (IASCC) of the core components. All of the candidate austenitic alloys evaluated at the University of Michigan have been shown to be susceptible to proton irradiation induced IASCC to varying degrees. The objective of this research is to improve understanding of the mechanism of IASCC of 316L stainless steel in supercritical water (SCW).

Two factors believed to influence the propensity for stress corrosion cracking are grain boundary sliding and localized deformation. This study will evaluate the dependence of these factors on the overall propensity of the grain to undergo slip as determined via the Taylor factor of the grain. The Taylor factor is a parameter that can be calculated from the crystallographic orientation of the grain, and a high Taylor factor indicates that more slip is required in the grain to accommodate the macroscopic deformation. Four tensile specimens of 316L stainless steel were irradiated with 2 MeV protons to a dose of 7 dpa at 400°C. Post-irradiation hardness measurements and beta counting measurements were performed to verify the samples were uniformly irradiated. The average irradiation induced hardening for the specimens was 237 Hv. The specimens will be incrementally strained in deaerated 400°C SCW to determine how cracking is influenced by the Taylor factor of the grain, and to determine if this relationship is a result of grain boundary sliding or localized deformation.

This research is supported by the U.S. Department of Energy under NERI grant DE-FC07-05ID14664.
Every year as part of the NERS 425 course, students participate in an experiment to determine the stoichiometry of a Ti$_x$N$_y$ sample using the reaction between a deuterium particle and a nitrogen nucleus: N$^{14}$(d,$\alpha$)C$^{12}$. Nuclear reaction analysis (NRA) is a well established surface analysis technique. In this method, an energetic particle (deuterium – produced by the Tandem accelerator at MIBL) interacts with the nucleus of an N atom (from the target) to give a reaction product ($\alpha$ particle) that can be measured. The students also use the backscattered yield from an RBS experiment to determine the amount of Ti in the sample by implementing simulation codes like RUMP or SIMNRA with the given experimental spectrum.

Prior to the start of the experiment, a short tutorial is given to the students on the accelerator, electronics, detectors, software, and vacuum components. After that, they work independently in a few groups with just the basic support from the MIBL staff, (required in the setup of the ion beam and the collection of the spectra). The students decide on a few parameters of the experiment and after that each group obtains spectra similar to the ones in the figure below.

![Typical NRA spectrum for the TiN film obtained during class.](image)

Conditions: beam energy: 1.4 MeV D$^+$,
solid angle 5 msr., detector angle 150°
FORMATION OF SURFACE AND EMBEDDED NANOPOROUS LAYERS IN GaSb USING HIGH ENERGY ION IRRADIATION

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In this work, we irradiate GaSb single crystals to investigate the effects of ion energy, mass, and fluence on the formation of ion irradiation-induced nanoporous structures. GaSb samples were irradiated with high energy Au\(^+\) ions in the range of 1-3 MeV and medium energy Kr\(^+\) ions of 150 keV. By varying the implantation conditions of the ions, we were able to produce structures with varying morphologies, as well as both surface and embedded porous layers.

Improving upon a proposed growth model for surface nanofibers, a qualitative universal growth model was developed to explain the presence and formation of the observed porous layers. Furthermore, methods for tailoring the properties of the porous layers through manipulation of the implantation conditions were developed. An additional model was proposed to account for surface layer removal effects observed after high ion fluence.

This study was supported by the U.S. Department of Energy under grants DE-FG02-02ER46005 and DE-AC05-76RL01830. Ion implantations were conducted in the Environmental Molecular Sciences Laboratory at PNNL and in the Michigan Ion Beam Laboratory at the University of Michigan, and analyses were completed at the University of Michigan’s Electron Microbeam Analysis Laboratory using equipment supported in part by the NSF under grants DMR-0320740 and DMR-9871177.

Cross-sectional SEM image of GaSb fiber layers irradiated with 1 MeV Au\(^+\) to 1 x 10\(^{14}\) ions/cm\(^2\), a). Insets show b) the surface layer, c) the presence of fully formed fibers, and d) a transition regime exhibiting a lower level of porosity.
RADIATION-INDUCED SEGREGATION AND PHASE STABILITY IN CANDIDATE ALLOYS FOR THE ADVANCED BURNER REACTOR

J. S. Penisten and G. S. Was, University of Michigan
J. T. Busby, Oak Ridge National Laboratory

The objective of this project is to study radiation-induced segregation (RIS) and phase stability in candidate alloys for application as structural materials in the advanced burner reactor. The alloys T91, HT9, and D9 have been irradiated with 2.0 MeV protons to doses of 3, 7, and 10 dpa at both 400°C and 500°C. T91 and HT9 are ferritic-martensitic alloys, while D9 is an austenitic alloy.

RIS measurements were performed by scanning transmission electron microscopy with energy dispersive x-ray spectroscopy (STEM/EDS). No pre-existing segregation was observed on any of the grain boundaries in any of the three alloys. Thus far, the T91 and HT9 specimens irradiated to 3, 7, and 10 dpa at 400°C have been analyzed. Alloy T91 exhibits Cr enrichment and Fe depletion at all three doses at 400°C, Figure 1. The magnitude of the Cr enrichment is ≤1.5 wt% at all boundaries studied. The point-to-point change in Cr concentration is inversely related in a nearly 1:1 ratio to the point-to-point change in Fe concentration. At all three doses at 400°C, T91 exhibits Si, Ni, and Cu enrichment. Alloy HT9 exhibits Cr enrichment at 3 dpa, but Cr depletion at 7 dpa and 10 dpa, Figure 2. However, the magnitudes of all observed Cr concentration changes are <1.0 wt%—less than the inherent uncertainty of the measurements. Ni enrichment by up to ~1.0 wt% is observed at all three doses in HT9 at 400°C.

Hardness measurements were performed before and after irradiation on all alloys. Irradiation-induced hardening is greater at 400°C than at 500°C, as expected. Hardening saturates around 7 dpa and the values compare well to those found in literature.

This work is supported by the U.S. Department of Energy under grant DE-FG07-07ID14828. A portion of this research was conducted at the SHaRE User Facility at Oak Ridge National Laboratory, which is sponsored by the Division of Scientific User Facilities, U.S. Department of Energy.

Figure 1. Grain boundary composition profiles in T91 following irradiation to 7 dpa at 400°C.
Figure 2. Grain boundary composition profiles in HT9 following irradiation to 7 dpa at 400°C.
Atom probe tomography (APT) has emerged as a complementary tool for understanding the IASCC mechanism by broadening our knowledge related to RIS, irradiation microstructure and localized deformation at an atomic scale. In this study, two alloys HP304 and HP304+Si were irradiated with 3 MeV protons at 360°C to 5 dpa. APT tips from both irradiated and unirradiated alloys were prepared using a focused ion beam (FIB) lift-out technique. The 3-d atom position and chemistry data were acquired using a local electrode atom probe (LEAP) microscope at the University of Alabama. The results show that solutes are uniformly distributed in the unirradiated alloys, however, after irradiation, nickel- and silicon-rich precipitates are formed in HP304+Si. Nickel was found enriched at grain boundaries and chromium was found depleted at grain boundaries for both irradiated alloys. The results are consistent with previous STEM/EDS measurements.

Support for this research was provided by EPRI.
VOID FORMATION IN He IMPLANTED Si

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\(^2\)Nuclear Engineering & Radiological Science Department, University of Michigan

Recently, there has been significant interest in the growth and thermal evolution of voids in Si by implantation of He because the voids in Si exhibit strong gettering of transition metals. Our understanding is that the void is created in two steps in bulk materials: the formation of small bubbles in the as-implanted state, and formation of a large void by subsequent thermal annealing.

A grand challenge is creating and assembling the voids on the semiconductor surface to construct functional structures. Here we report the void formation on the surface of Si by removing the amorphous layer using a focused ion beam on He implanted Si. High flux and high fluence were used to form large bubbles without annealing. At peak position where the concentration of He is the largest, a non-spherical shaped bubble formed with size around 50 nm, while at a deeper position, a spherical bubble with 2 nm in diameter was formed and was uniformly distributed. The high dose, high dose rate can be responsible for this phenomenon. This method opens up a promising new approach to the application of these voids in Si technology including, for example, nanoscale photonic and electronic devices.

This work was supported by the Office of Basic Energy Science of the U.S. Department of Energy through Grants No. DE-FG02-02ER46005.

(a) TEM cross-section view showing the variation of bubble size with the implantation depth.
(b) TEM plan view at the area with low concentration of He showing the small bubble distribution (diameter of ~4 nm).
(c) SEM images showing the void formation at the peak position of He after removing the amorphous layer by focused ion beam. Energy 160 keV, dose \(5 \times 10^{17} \text{cm}^{-2}\), dose rate \(2.5 \times 10^{13} \text{cm}^{-2} \text{s}^{-1}\), at room temperature.
During October 2008 over the week of 12-17, Michigan Ion Beam Laboratory (MIBL) hosted the 42nd Symposium of North Eastern Accelerator Personnel (SNEAP). This was the first time that MIBL was involved as an organizer in this international event.

SNEAP – founded in 1968 - is not just a yearly forum but also an entity which has a strong presence in cyberspace as users and accelerator operators come together online to ask questions and receive answers related to the construction, functionality and operation of particle accelerators. The SNEAP community meets annually to discuss and exchange information for the benefit of all present. The topics covered include ion sources, electrostatic accelerators, telemetry and control systems, cryogenic systems, accelerator safety issues and many other subjects relevant to the operation of small to large sized electrostatic accelerators. The meeting format includes submitted papers, laboratory reports, contributed papers and open discussions. Space for vendors is made available to permit attendees to speak directly with manufacturers and their representatives and to inspect many new devices and products. This year’s attendees include representatives from universities, National Laboratories, and Industrial Research Laboratories from seven countries: US, Australia, Canada, France, Germany, Italy and Finland.

The participants got the chance to visit the National Superconducting Cyclotron Laboratory (NSCL) at the Michigan State University and, in a more relaxed environment, the Henry Ford Museum and Greenfield Village as landmarks of the area. MIBL staff also arranged, with help from the Nuclear Engineering department, visits to the major labs on North Campus of the University of Michigan.

The participants had many ideas exchanged in a friendly environment and attended a good number of presentations pertinent to their work. Of significance this year was the fact that during each morning of the conference, a workshop was organized targeting a larger audience. The topics were: “Application Trends for Electrostatic Accelerators” by Greg Norton (NEC), “Ion Sources” by Greg Norton (NEC) and Frank Chmara (Peabody Scientific), “Electrostatic Accelerators” by David Weisser (ANU), and “HV and Radiation Safety” by Martha Meigs (ORNL). Considering the positive input from all the participants, we believe that this year’s event was a success.
INTRINSIC STRESS EVOLUTION IN NANOCRYSTALLINE DIAMOND THIN FILMS WITH DEPOSITION TEMPERATURE

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Nanocrystalline diamond (NCD) films possess excellent mechanical properties, low surface roughness, tunable electrical conductivity, biocompatibility, and a wide electrochemical potential window. These properties in turn provide NCD many potential applications in industry. However, the residual stress generated in the film is complicated, which could limit the reliability of NCD based devices. Therefore, it is critical to control the stress for desired applications. In this study, we investigated the stress evolution in NCD films deposited at different temperatures (from 400°C to 800°C). Results showed that the intrinsic stress gradually changed from tensile to compressive with decreasing deposition temperature. Most importantly, the intrinsic stress can be tailored to zero by adjusting the deposition temperature, which is critical to many applications. It has been proved that more H as well as sp²-bonded carbon was incorporated into the grain boundaries, which was responsible for the evolution of stress and other mechanical properties with deposition temperature (Figure 1). Moreover, all the NCD films showed excellent mechanical properties (Figure 2).

Figure 1. The temperature dependence of H content during film formation.

Figure 2. The temperature dependence of Elastic Modulus for the NCD films.
RADIATION INDUCED MICROSTRUCTURE, DISLOCATION CHANNELLING AND IODINE-INDUCED STRESS CORROSION CRACKING (I-SCC) IN PROTON-IRRADIATED ZIRCALOY-4

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G. S. Was, University of Michigan

The radiation-induced microstructure, strain localization, and iodine-induced stress corrosion cracking (I-SCC) behavior of recrystallized Zircaloy-4 proton-irradiated to 2 dpa at 305°C was examined. <a> type dislocation loops having 1/3<11\bar{2}0> Burgers vector and a mean diameter and density of respectively, 10 nm and 17×10^{21} m^{-3}, were observed while no Zr(Fe,Cr)\textsubscript{2} precipitate, amorphization or Fe redistribution were detected after irradiation. After transverse tensile testing to 0.5% macroscopic plastic strain at room temperature, almost exclusively basal channels were imaged. Statistical Schmid factor analysis shows that irradiation leads to a change in slip system activation from prismatic to basal due to a higher increase of critical resolved shear stresses for prismatic and pyramidal slip systems than for the basal slip system. Finite element calculations suggest that dislocation channeling occurs in the irradiated proton layer at an equivalent stress close to 70% of the yield stress of the irradiated material, i.e. while the irradiated layer is still in the elastic regime for a 0.5% applied macroscopic plastic strain. Comparative constant elongation rate tensile tests performed at a strain rate of 10^{-5} s^{-1} in iodized methanol solutions at room temperature on specimens both unirradiated and proton-irradiated to 2 dpa demonstrated a detrimental effect of irradiation on I-SCC.

Support was provided by CEA DSOE Basic Research.

(a) TEM micrograph of <a> dislocation loops at g = 1 0 \bar{1} 1.
(b) Dislocation loop diameter distribution in Zircaloy-4 proton-irradiated to 2 dpa at 305 °C.
SURFACE CHARACTERIZATION OF FEMTOSECOND LASER MODIFIED MATERIALS

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The experiments performed at the Michigan Ion Beam Lab mainly focused on surface characterization of various laser modified metals. 4330 titanium modified steel, CMSX-4 nickel base superalloy, and AXJ-530 magnesium were laser machined using a Clark-MXR Ti-sapphire femtosecond laser operating at 780 nm, 1000 Hz, and 800 \( \mu \)J pulse energy. Surface characteristics, such as average roughness and material removal depth, were measured using the Michigan Ion Beam Lab’s Dektak 3 Profilometer. This machining investigation required a method for verifying predicted laser removal rates. By using the profilometer we were able to determine the post machining surface depths and compare them to theoretical values. Shown in the following representative figure, the depth of the machined areas can be determined by comparing the original surface height with that of the machined area.

This work was supported by the Dept. of Navy/DARPA under Grant # F013434.
CONTROLLING NANOPARTICLE DISTRIBUTION IN THIN FILM POLYMER MATRIX

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In this work, gold nanoparticles capped with ligands are incorporated into a chemically identical polymer matrix. Depending on the core size, chain length of ligands and the chain length of the polymer hosts, particle distribution can be controlled by changing the miscibility between the particle and the polymer host. The RBS analysis was performed at MIBL. The figure below shows the RBS spectrum for the polymer nanocomposite film on the silicon substrate overlapped on the simulated spectrum. By simulating the experimental data with the given film thickness (92 nm) and total concentration (2 wt %) of particles, it was possible to get nanoparticle concentration as a function of the depth with 10% precision. From the experimental and simulated result, particles are distributed throughout the film with slightly more packing on the substrate and on the free surface. Despite the noise in the experimental data, we were able to divide the experimental data into six layers and determine the concentration of Au, which was reasonably close to the SIMS analysis. On one hand, when particle size is small (4-5 nm; core + ligand size), there is no entropic loss to mix into polymer hosts. Conversely, when particle size is large (10-11 nm) with similar ligand size, particles will be immiscible to polymer hosts. In this case host chains need extra stretching energy to conform the particles; thus particles will likely be segregated to the free surface and to the substrate to minimize the energy loss. By controlling the distribution of incorporated nanoparticles, it would be possible to enhance some mechanical and optical properties of the material.

Experimental spectrum (red) and simulated spectrum (blue) for Au particles implanted in the polymer film on a Si substrate.
INFLUENCE OF IMPLANTATION TEMPERATURE ON BLISTERING IN GaAs:N

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Ion-implantation-induced blistering in semiconductors has mainly focused on light ion (H or He) implantation into silicon and GaAs. In those cases, the blister formation process is apparently implantation-temperature-dependent, with substantial differences in damage profiles and exfoliation depths for low- and high-temperature implantations. We have investigated the influence of implantation temperature on blister formation in GaAs:N layers produced by N-implantation into GaAs followed by rapid thermal annealing. Scanning electron microscopy (SEM) of the implanted-plus-annealed GaAs:N layers reveals blistered surfaces with similar feature sizes and densities, independent of implantation temperature. Atomic force microscopy (AFM) reveals similar popped blister (crater) depths in each case, suggesting a consistent depth of bubble formation. To determine the depth-dependence of lattice disorder, we utilized channeling-Rutherford backscattering spectrometry (RBS) with a 2 MeV He++ beam. Similar depth-dependencies of the normalized yield are observed, suggesting similar post-annealing damage distributions, independent of annealing temperature. The implanted-plus-annealed layers exhibit local maxima with normalized yields similar to that of the random GaAs yield. Interestingly, these localized maxima occur at depths similar to the crater depths revealed by AFM. Thus, it is likely that this local maximum in yield is due to dechanneling of He++ at the bubbles.  

Figure 1. SEM/AFM images collected from the same regions of the surfaces of (a)/(b) low-temperature implanted and (c)/(d) high-temperature implanted-plus-annealed GaAs:N. The solid and dashed circles in (a) and (c) reveal ~2 µm circular blisters and craters, respectively. The line-cut profiles in (b) and (d) reveal crater depths of ~200 nm.

This work was supported in part by the National Science Foundation under grant CMMI-0700301, and graduate fellowships from the Michigan Memorial Phoenix Energy Institute and NSF.

Figure 2. Channeling-RBS spectra as a function of backscattered energy for low- and high-temperature implanted-plus-annealed GaAs layers, both in comparison with GaAs in the random and (001) aligned configurations. The local maxima at depths corresponding to the crater depths, with normalized yields similar to that of the random GaAs yield, are likely due to dechanneling by bubbles.
AN INITIATION STUDY OF PROTON IRRADIATED AUSTENITIC STAINLESS STEELS

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The objective of this research is to perform stress corrosion initiation tests on these proton-irradiated specimens exposing them to PWR primary water under constant load and subsequently performing SEM in-situ CERT tests to characterize the initiation precursor.

In this work, a first proton irradiation was conducted to 5 dpa at 360°C using 2 MeV protons at the Michigan Ion Beam Laboratory. Due to the shape and thickness (very thin - .5 mm) of the samples this irradiation proved to be very challenging but was carried on successfully (Figure below).

Experimental work is in progress to analyze the irradiated samples.

![Sample geometry for the first irradiation.](image1.png)

Figure 1. Sample geometry for the first irradiation.

![Stage loaded with the samples during irradiation.](image2.png)

Figure 2. Stage loaded with the samples during irradiation.
ION BEAM INDUCED THIN LAYER EXFOLIATION

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The purpose of the research is to find the differences in blistering parameters due to the differences in implantation temperature. The implantations were performed at liquid nitrogen cooled, water-cooled and un-cooled substrates.

The idea was to find which condition was better for exfoliation depending the desired annealing temperature and time. The results are not conclusive yet. For a future project, perhaps the differences in implantation dose would also be studied.

The implantations were done at the Michigan Ion Beam Laboratory, using H⁺ ions with energy of 115 kV and 130 kV and a dose rate of 1x10¹⁶ At/cm²/h.

Table 1. Irradiation matrix.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Target</th>
<th>Dose</th>
<th>Energy</th>
<th>Temperature</th>
<th>Beam Current</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si1</td>
<td>4” Si</td>
<td>4.0 E16 H⁺/cm²</td>
<td>115 keV</td>
<td>Room</td>
<td>100-150 μA</td>
</tr>
<tr>
<td>Si2</td>
<td>4” Si</td>
<td>8.0 E16 H⁺/cm²</td>
<td>115 keV</td>
<td>Room</td>
<td>100-150 μA</td>
</tr>
<tr>
<td>Si3</td>
<td>4” Si</td>
<td>8.0 E15 H⁺/cm²</td>
<td>115 keV</td>
<td>Room</td>
<td>100-150 μA</td>
</tr>
<tr>
<td>Si4</td>
<td>4” Si</td>
<td>4.0 E16 H⁺/cm²</td>
<td>130 keV</td>
<td>-196°C</td>
<td>100-150 μA</td>
</tr>
</tbody>
</table>
Pyrolytic carbon (PyC) has many potential applications due to its superior mechanical hardness, bio-compatibility, high thermal conductivity, etc. For example, PyC has been proposed as one of the cladding material candidates in advanced nuclear power concepts. As a cladding material, it needs mechanical robustness and the ability to retain nuclear reaction products (such as silver). In addition, it is noteworthy that PyC with implanted Ag has high bactericidal rate according to recent studies, a property that has some promising applications in modern medical science. PyC samples were prepared by chemical vapor deposition at a temperature of 1300°C. The silver implantation was conducted at the Michigan Ion Beam Laboratory (MIBL). The dose and ion energy were: $2.0 \times 10^{16}$ cm$^{-2}$ and 400 keV, respectively. The implantation temperatures were set at room temperature and at 300°C.

PyC has a-b plane isotropy and anisotropy along the c plane. This anisotropy along the c plane was partially broken after Ag-plantation. Meanwhile the smooth surface in the original PyC changed to near-periodic cellular clusters after bombardment. Chemical bonding revealed by EELS showed that sp$^2$ bonding (graphite) in original PyC was partially broken during Ag implantation. The implanted Ag ions were agglomerated around the cellular clusters, and formed Ag particles with FCC structure. Stacking faults and nano-twins were observed in FCC Ag particles. Accordingly, we believe that displacement spike with silver agglomeration leads to the morphology changes of PyC surface.

This work has been supported by U.S. Department of Energy BES under Grant No. DE-FG02-02ER46005.

SEM micrograph of PyC (a) before Ag-implantation, (b) after Ag-implantation.
The controlled formation of semiconductor nanocomposites offers a unique opportunity to tailor functional materials with a variety of novel properties. In particular, nanocomposites consisting of InAs nanostructures embedded in GaAs have been proposed for high efficiency photovoltaics and high figure-of-merit thermoelectrics. A promising approach to nanocomposite synthesis is matrix-seeded growth, which involves ion-beam-amorphization of a semiconductor film, followed by nanoscale recrystallization via annealing [1]. In earlier studies of In+ implantation into GaAs, it has been suggested that In aggregates upon annealing [2-4], suggesting the possibility of selective formation InAs-rich nanocrystals in a GaAs matrix. To date, the nanostructure of In+ implanted plus annealed GaAs has not been reported.

In this work, we are endeavoring to fabricate InAs-rich nanocrystals within a GaAs matrix, using a combination of blanket and focused ion beams. For blanket ion implantation, we used the NEC ion implanter at MIBL with an indium tin oxide source. During implantation, the samples were maintained at room temperature via water cooling. To “simulate” focused-ion-beam, the ion energy was set at 30 keV, with ion doses ranging from 1 to 8 x 10\(^{16}\) cm\(^{-2}\).

TEM inspection of the GaAs:In samples suggests the formation of a thin damaged surface layer (Figure 1). Indeed, the maximum obtainable In concentration is predicted to be limited by the high GaAs:In sputter yield. Presumably, the resulting limited GaAs:In incorporation hinders the formation of nanocrystals. We are currently endeavoring to increase the implanted In concentration by utilizing a thin “sputter mask”, i.e. a layer with In sputter yield lower than that of GaAs. For example, profile code simulations of a SiO\(_2\)/GaAs structure suggest an increase in maximum (implanted) In concentration in the GaAs by more than an order of magnitude (Figure 2).

PUBLICATIONS AND PRESENTATIONS

Publications


Presentations


G. S. Was, “Emulating Neutron Irradiation Damage with Ions – when the Simulation Tool is Better than the Real Thing,” Grand Challenges in Ion Beam Research – AAMU/Huntsville 20$^{th}$ Anniversary, Huntsville, AL, August 2008.


