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Gallium Interactions with Zircaloy

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AMARILLO NATIONAL RESOURCE CENTER FOR PLUTONIUM/
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Final Thesis Report

Gallium Interactions with Zircaloy

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Abstract

High fluence ion implantation of Ga ions was conducted on heated Zircaloy-4 in the range of 10^{16} - 10^{18} Ga ions/cm². Surface effects were studied using SEM and electron microprobe analysis. The depth profile of Ga in the Zircaloy was characterized with Rutherford backscattering and SIMS techniques. Results indicate that the Zirc-4 is little affected up to a fluence of 10^{17} Ga

ions/cm². After implantation of 10^{18} Ga ions/cm², sub-grain features on the order of 2 μ m were observed which may be due to intermetallic compound formation between Ga and Zr. For the highest fluence implant, Ga content in the Zirc-4 reached a saturation value of between 30 and 40 atomic %; significant enhanced diffusion was observed but gallium was not seen to concentrate at grain boundaries.

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To: S, K, T, and R

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1. INTRODUCTION

An important option for the disposition of weapons-grade plutonium (WGPu) in the United States is the conversion of weapons-grade plutonium into mixed-oxide (MOX) reactor fuel. MOX fuel fabricated in this way must be compatible with currently used nuclear fuel components. Since US WGPu contains small amounts of gallium (about 1% by weight) (Toevs, Beard), it is necessary to study the effects that gallium may have on fuel components such as reactor fuel cladding (Lindemer, 1995; Wilson, DiStefano, King, Manneschmit, and Strizak, 1997, 1997; Wilson, et al, 1997). The study presented here is a step toward investigating some of the effects that gallium may have on zircaloy cladding during reactor operation.

As a result of the reprocessing of spent fuel used in European nuclear programs, many studies have been conducted on the production and behavior of MOX fuel in traditional reactors (MacLeod and Yates, 1993; Krellman, 1993; Paratte and Chavla, 1995). However, only a few studies have examined the effect that MOX fuel containing WGPu may have in a reactor (JEOL, Ltd, 1991). Presently, studies investigating the interaction of zircaloy cladding materials with gallium are being conducted by Oak Ridge National Laboratory (Wilson, DiStefano, King, Manneschmit, and Strizak, 1997; Wilson, et al, 1997). These studies have primarily focused on the mechanical properties of heated zircaloy cladding exposed to gallium, paying close attention to

corrosion and intermetallic compound formation.

MOX fuel fabricated using the "ARIES" process will probably contain gallium in the elemental or oxide (i.e. Ga_2O_3) form and will probably be present in the range of 10 ppm by weight (Wilson, et al, 1997). The high temperatures and severe irradiation environment present in a reactor may lead to diffusion of this gallium into the fuel cladding. It is estimated that if one ppm of Ga is present in MOX fuel, and all of this Ga diffuses into the adjacent cladding, the zircaloy would be impacted by roughly 2.4×10^{16} Ga atoms/cm² (Hart, et al, 1997).

This study focuses on the effects of gallium ion implantation into zircaloy cladding material to investigate the effects that gallium may have in a reactor. To simulate these effects, Zircaloy targets maintained at a typical cladding temperature were implanted with high fluences of Ga to both introduce Ga and to approximate the damage produced by fission fragments in a reactor environment. Ion implantation provides a more accurate representation of the irradiation environment in a reactor than merely thermally introducing Ga into Zircaloy. The effects of Ga implantation are observed by closely examining the microstructure of the implanted area. The techniques used to examine the implanted samples focus on determining the surface effects, lateral distribution, and depth profile of gallium in the implanted zircaloy.

2. EXPERIMENTAL

2.1 EXPERIMENTAL OVERVIEW

The experimental procedure for the proposed study has two goals: ion implantation of gallium into zircaloy cladding and subsequent analysis of the microstructure of the implanted samples. Figure 1 shows an overview of the basic steps and methodology used for the experiment. Sample preparation at the University of Texas was followed by ion implantation at Texas A&M University. High fluence ion implantation of 100 keV gallium in the range of 10^{16} - 10^{18} Ga ions/cm² was conducted on Zircaloy-4 targets maintained at a typical cladding temperature of 375°C.

Figure 1 also shows the analysis techniques important to the experiment and their roles in examining implanted samples. Surface morphology of the implanted areas is examined by using scanning electron microscopy (SEM). Electron Microprobe analysis (EM) investigates the lateral distribution of Ga in the Zircaloy. X-ray diffraction techniques were employed to identify the formation of possible intermetallic compounds between Ga and Zr. Finally, Rutherford Backscattering (RBS) and Secondary Ion Mass Spectrometry (SIMS) were used to determine the depth profile of implanted Ga in the Zircaloy cladding material.

Figure 1: Flowchart of Basic Project Steps

2.2 ACCELERATOR DESCRIPTION

The ion beam irradiations were performed using the 200 keV accelerator system in the Ion Beam Laboratory at Texas A&M University. A general layout of the accelerator is shown in Figure 2. The accelerator consists of three main components: the source, the beam line, and the target chamber, each of which is described below.

2.2.1 Source

The source for the 200 keV accelerator used in this study is a Danfysik model 911A ion source. The source is a hot filament, hollow cathode source that can accommodate both gaseous and solid source materials. The ion source is made up of a tungsten filament, a tantalum anode, and an oven. A high current is passed through the tungsten filament causing thermionic emission. The voltage between the cathode and anode accelerates emitted electrons causing ionization of the gas in this region of

the source. The oven consists of a boron-nitride insulated cylinder wrapped with a tantalum oven wire and provides for evaporation of solid source materials. For an appropriate combination of gas pressure, filament current, and anode voltage, a plasma is formed in the hollow cathode region of the source. Ions are then extracted from the source by the high voltage extraction and focusing elements and then accelerated down the accelerating column.

2.2.2 Beam Line

The beam line consists of various components that shape and provide additional focusing of the desired ion beam. A large magnet capable of fields up to 8000 Gauss provides for isotopic separation. As the magnetic field is adjusted, the ions in the beam are deflected at an angle proportional to their charge to mass ratio. The beam is deflected down the beam line using horizontal and vertical deflection plates and the quadrupole acts to focus the beam. In this

way, the desired ions can be deflected down the beam line to the target chamber. Finally, the beam is passed through a set of collimators to achieve a beam of desired size. The collimators consist of two sets of plates with different diameter holes from 1/32" to 5/8".

2.2.3 Target Chamber

The target chamber is a large cylindrical volume approximately 40 cm in diameter and 35 cm high equipped with many instrumentation ports and connections. A schematic of the target chamber is shown in Figure 3. The target chamber pressure is maintained at approximately 1×10^{-7} torr by cryogenic and ion pumps. Figure 3 shows the location of the Zirc-4 sample to be implanted. The Zirc-4 target is mounted on a goniometer that can be used to move the target or align it with the beam. The target holder is electrically connected to a current integrator that provides an accurate current measurement of the implanted ions. For the heated implants in this experiment, the target is surrounded by a heating assembly. The heater assembly is discussed in more detail below. Finally, a PIPS surface barrier type detector is part of a particle detection system

that can be used to conduct Rutherford Backscattering (RBS) experiments (see Figure 3).

2.2.4 Heater Assembly

To maintain the Zirc-4 target at a high temperature, it is surrounded by the heater assembly or oven shown in Figure 4. The oven is composed of three cylindrical shells. The outer layer is a molybdenum sheet that minimizes heat loss. The central layer consists of a quartz cylinder wrapped with a stainless steel heater wire. Current is supplied to the heater wire by an external power supply. For a more accurate beam current reading, The inner stainless steel cylinder of the oven is used as a Faraday cup to suppress secondary electrons generated during ion irradiation.

2.2.5 Detection System

Rutherford Backscattering analysis of the sample in the target chamber is carried out with the detection system shown in Figure 5. A biased Canberra PIPS surface barrier detector detects backscattered light ions producing an output pulse height proportional to the energy of the particle. Detector bias is supplied by an Ortec 428

Figure 2: Overview of the 200 keV Accelerator System

Figure 3: Schematic of the Target Chamber Used for Ion Implantation and RBS Analysis

power supply. The output signal from the detector is sent through a Canberra 2001A preamplifier and Canberra 2020 amplifier to a Series 35+ Canberra multi-channel analyzer (MCA) to store the backscattered energy spectrum. A pulser signal to the preamp input is supplied by a Canberra 807 Pulser to adjust the amplifier gain and check the electronic resolution of the system. Electronic noise levels for $^3\text{He}^{++}$ detection was about 3.0 keV.

2.3 EXPERIMENTAL PROCEDURE

Several small (1cm diameter x 0.5mm thick) discs of Zircaloy 4 were prepared by the University of Texas at Austin for ion implantation. The discs were polished to a mirror-like finish using mechanical polishing techniques. The samples were first polished

with silicon paper with successive grades down to 1200 grit. Next, diamond suspension polishing with a polishing cloth was utilized until no visible marks were left on the sample. Finally, polishing with colloidal silica was conducted until optical microscope examination showed no scratches.

Solid Ga (99.999% pure) material was used as the source material in the ion source. Approximately 0.1 g of Ga was loaded into a small cylindrical quartz crucible and placed inside the source oven. To initiate a plasma discharge in the source, argon was supplied to the source through the gas inlet by a leak valve. Next, the source oven current was slowly increased as the argon flow was decreased.

Figure 4: Heater Assembly used for Ga Implantation [10].

Figure 5: Detection System Used for RBS Analysis

The two isotopes of gallium, ^{69}Ga and ^{71}Ga , were observed using the beam profiler. Because of the larger ratio of ^{69}Ga to ^{71}Ga , ^{69}Ga was chosen to conduct the implants and minimize the implantation time. A magnet setting of 6790 Gauss was used to deflect a beam of 100 keV ^{69}Ga .

The first set of gallium implantation was conducted on Zirc-4 samples, which were not heated and remained at room temperature. The first purpose of room temperature implantation goal was to achieve a high-fluence gallium beam. The second was to discover what fluence of gallium ions would cause a noticeable effect on the Zircaloy and have a measurable depth profile using Rutherford Backscattering techniques. These trial runs showed that implantation below 10^{16} Ga ions/cm² was completely unobservable using RBS and SEM techniques. The trial also showed that a 1 μA ^{69}Ga beam could be achieved using this system.

For gallium implantation into heated targets, the Zirc-4 samples were mounted on the goniometer, and the heater assembly shown in Figure 4 was placed around the target. Temperature inside the heater was monitored closely using two K-type Chromel-Alumel thermocouples inside the target chamber. Figure 6 shows the setup used during ion implantation. The first thermocouple was mounted to the back of the sample for accurate measurement of the Zircaloy target temperature. Thermocouple 2 was positioned against the goniometer motor nearest to the heater assembly. The purpose of this thermocouple was to prevent overheating of the goniometer motor above 100°C. In addition, braided copper straps were wrapped around the goniometer motors and connected to the cold plates in the target chamber.

Figure 6: Target Chamber Configuration for Heated Ga Implantation.

Current was supplied to the heater assembly wire using a Sorenson DCR40-60A DC power supply. Current was slowly increased until the target had the desired equilibrium temperature. A setting of 2.5 amps maintained the target current at the required temperature of 375°C (a reading of 15.34 Volts from the thermocouple). During operation of the heater, the voltage of thermocouple 2 did not exceed 3.89 V or 95°C. After the target reached the operating temperature of 375°C, ion implantation of 100 keV Ga ions was performed. Table 1 shows a summary of the ion implantation data.

Table 1: Accelerator Settings for Gallium Implantation

Accelerating Voltage	100 kVolts
Beam Line Pressure	6-8 μ torr
Magnet Setting	6790 Gauss
Filament Current	25 Amps
Source Oven Current	15 Amps
Anode Voltage	60 Volts
Anode Current	1.5 Amps

To give a relatively large implantation spot and have a reasonable implantation time, the 1/8" collimator was used for the gallium

implantation. A ^{69}Ga beam of approximately 1 μA was measured on the target through a 1/8" collimator. Three implantation spots were made on the Zirc-4 target in the arrangement shown in Figure 7. The first implant was performed slightly off center and the goniometer and sample were rotated 90 degrees between each subsequent implant (see Figure 7). Target current was measured by an Elcor A310C current integrator that gave a direct current reading on the target and an integrated total charge implanted. A -200 V bias was applied to the inner heater cup to suppress secondary electron emission. Table 2 shows the current integrator readings for each implantation that corresponds directly to the amount of gallium introduced to the Zircaloy.

Table 2: Current Integrator Measurements for Heated Gallium Implantation

Implant Goal (ions/cm ²)	Current Integrator Reading (Coulombs)
10^{16}	1.2707×10^{-4}
10^{17}	1.2685×10^{-3}
10^{18}	1.2683×10^{-2}

Figure 7: Diagram Showing the Implanted Areas on the Sample

2.4 SAMPLE ANALYSIS

Following the heated implantation, the sample was allowed to cool to room temperature for Rutherford Backscattering analysis. The solid state PIPS detector located at 160 degrees shown in Figure 4 was used to detect backscattered ^3He ions. The backscattered energy spectrum of doubly-ionized 280 keV ^3He ions was measured with the detection system shown in Figure 5. After observing approximately 0.5 nA of $^3\text{He}^{++}$ through a 1/16" aperture, three different areas were analyzed: the $10^{17}/\text{cm}^2$, $10^{18}/\text{cm}^2$, and an unimplanted area. Table 3 shows the current integrator readings for the charge collected during RBS analysis of the Zirc-4 sample.

Table 3: Current Integrator Readings for RBS Analysis

Area	Implanted Charge (Coulombs)
$10^{17}/\text{cm}^2$	100.10×10^{-8}
$10^{18}/\text{cm}^2$	100.02×10^{-8}
Unimplanted	100.09×10^{-8}

Following the Rutherford Backscattering, the Zirc-4 sample was analyzed using a variety of techniques. To examine the surface features of the implanted Zircaloy, scanning electron microscopy (SEM) of the sample was conducted at the Electron Microscopy Center at the Texas A&M Biology Department using the JEOL JSM-T330A Scanning electron microscope. Because the Zirc-4 samples were conductive, no sample preparation was needed before using the SEM. Initial trials with the SEM showed that an accelerating voltage of 15 kV gave detailed images for magnifications of up to 5000 times. SEM micrographs of implanted and unimplanted areas were taken at 2000X and 5000X.

Next, the sample was examined using the Cameca SX-50 Electron Microprobe located in the Texas A&M Geology Department. The microprobe is equipped with a wavelength-dispersive spectrometry system (WDS) capable of detecting x-ray emission from elements of atomic number five or greater. The microprobe is equipped with optical and backscattered electron (BSE) microscopes together with the WDS capabilities. The microprobe was used to study the lateral distribution of gallium in the implanted Zircaloy by conducting x-ray maps of the implanted areas as well as a linear scan across the sample.

To identify possible intermetallic phase formation, X-ray diffraction was then conducted using an x-ray diffractometer with a rotating 2-Theta detector setup and Cu $K\alpha$ x-rays. Two diffraction spectra were obtained using a 1/6th degree slit. Due to the relatively small size of the sample and implanted areas, a nickel filter was added to the 1/6th degree slit to attempt to collimate the x-ray beam on the highly implanted area. The first spectrum was an analysis of the implanted sample using a nickel filter. Also using the nickel filter, the second spectrum was obtained from an unimplanted Zirc-4 sample, which was used as a standard for comparison.

Finally, to supplement the Ga depth profile data obtained from RBS measurements, secondary ion mass spectrometry analysis was conducted by Charles Evans and Associates (SIMS Analysis). The center of each implanted (10^{16} , 10^{17} , $10^{18}/\text{cm}^2$) area was analyzed for gallium content by sputtering the surface to a depth of over 2000 Å.

3. RESULTS AND DISCUSSION

3.1 SURFACE MORPHOLOGY

Surface morphology of the implanted Zircaloy-4 was examined using the JEOL JSM-T330A scanning electron microscope, which has a resolution on the order of 5 nm (JEOL, Ltd, 1991). Figure 8 shows an SEM micrograph obtained from an area near the center of the Zirc-4 sample. It is a secondary electron image taken of an unimplanted area of the sample after heating and ion implantation on other areas has occurred. The magnification shown in Figure 8 is 5000X, and the micrograph appears very similar to the as-polished sample which is quite flat and featureless. The small pits in the lower left hand corner of Figure 8 were used to aid in focusing since the surface is flat.

Next, Figure 9 shows another SEM micrograph at 5000X. The image in Figure 9 is a micrograph of the area implanted with 10^{16} Ga ions/cm². As is shown in Figure 9, gallium implantation at the fluence of 10^{16} ions/cm² appears to have little or no effect on the surface of the sample. The scale bar noted on this figure is 1 μ m. Also Figure 9 shows a black arrow drawn across one of the typical grains in the Zirc-4 sample. The grain size of the Zirc-4 sample is seen to be from 10-20 μ m. Again, the pit in the center of this micrograph was used as a feature on which to focus.

A micrograph of the 10^{17} /cm² area at 5000X is pictured in Figure 10. It can now be seen that the surface appears rough, possibly due to increased sputtering of the surface from the higher fluence gallium implantation. However, up to a fluence of 10^{17} Ga ions/cm², virtually no observable effects such as cracking, pitting (Cuomo and Rossnagel, 1989), or blistering are seen.

Figure 11 shows a dramatic effect on the surface structure for the 10^{18} ions/cm² implant. As seen in the 5000X photo of Figure 11, a grain-like structure has formed

after the highest fluence implantation. As the size of the scale bare on Figure 11 indicates, the size of these grains is on the order of 2 μ m. The granular structure developed after the 10^{18} /cm² implant is also shown in Figure 12 at 2000X. It should be noted when comparing Figure 11 to Figure 9 that these 2 μ m sub-grains do not correspond to grains in the fabricated Zirc-4 sample, which are on the order of 10-20 μ m.

3.2 LATERAL DISTRIBUTION OF Ga

Several analytical techniques were conducted using the electron microprobe. A backscattered electron (BSE) image of the area implanted with 10^{18} Ga ions/cm² is shown in Figure 13. Operating conditions used to obtain this figure were an accelerating voltage of 20 keV and a beam current of approximately 3 nA. Like the SEM micrograph of Figure 11, Figure 13 is an approximately 5000X image and shows that a granular structure on the order of 2 μ m is present. It should also be noted that lighter and darker areas in the BSE image suggest that regions of different average atomic number may be present (areas of higher atomic number appearing lighter in the image) (Reed, 1993), thus the darker areas between these sub-grains suggest a higher Ga concentration. The very dark spots seen in Figure 13 are due to the presence of surface blemishes present before implantation.

The lateral gallium distribution in the highly implanted area was investigated using the x-ray mapping capabilities of the microprobe. Figure 14 shows an x-ray map of the Ga K α and L α x-rays superimposed upon a 10,000X BSE image of the same area on the 10^{18} Ga/cm² implantation spot. The x-ray map of Figure 14 shows that the Ga concentration is not distributed uniformly, and suggests, like Figure 13, that the Ga concentration seems to be more concentrated between these 2 μ m sub-grain features; this

Figure 8: SEM Micrograph of an Unimplanted Zirc-4 Region

Figure 9: SEM Micrograph of Zirc-4 Implanted with 10^{16} Ga ions/cm²

Figure 10: SEM Micrograph of Zirc-4 Implanted with 10^{17} Ga ions/cm²

Figure 11: SEM Micrograph of Zirc-4 Implanted with 10^{18} Ga ions/cm²

Figure 12: SEM Micrograph 2000X of Zirc-4 Implanted with 10^{18} Ga ions/cm²

Figure 13: BSE Image of Area Implanted with 10^{18} Ga ions/cm²

may suggest that presence of Ga-Zr phases forming in the implantation areas. It should also be noted that the effective spatial resolution of the microprobe at the energy of 20 keV is on the order of $1\text{ }\mu\text{m}$ because of the large electron interaction volume (Goldstein, 1992). An attempt to decrease the beam energy to 5 keV and decrease the size of the interaction volume was made with little success because of the poor focusing capabilities of the microprobe at this lower energy.

A line scan across a section of the 10^{18} Ga/cm^2 implant area was conducted using the microprobe. Figure 15 shows a BSE image of the 10^{18}/cm^2 implant spot which is an approximately 13,000X image (note the scale bar). Figure 16 shows the results from a line scan across the same area of the high fluence implant area as shown in Figure 15. The optical microprobe and sensitive stage were used together to conduct a $6\text{ }\mu\text{m}$ line scan across the region shown as the black arrow in Figure 15. A beam energy of 10 keV and beam current of 3.0 nA were used during the scan. Figure 16 is the result of counting the Ga $K\alpha$ x-rays emitted at each $0.2\text{ }\mu\text{m}$ step as the beam was scanned across the region. The counts shown in the y-axis of Figure 16 are the raw x-ray counts collected for 30 seconds at each $0.2\text{ }\mu\text{m}$ step of the scan. Figure 16 shows, like the x-ray map of Figure 14, that the gallium concentration is not uniform over the surface of the implanted area, which again may indicate possible Ga-Zr compound formation. In fact, it is seen that the Ga concentration suggested by the line scan may differ laterally by as much as 30% over regions of the 10^{18}/cm^2 implant. It should again be noted that the interaction volume of 10 keV electrons (and therefore the volume of excited x-rays) used in the scan is slightly greater than $0.5\text{ }\mu\text{m}$, which limits the spatial resolution of the detected x-rays in Figure 16.

3.3 DEPTH PROFILE ANALYSIS

Rutherford backscattering analysis was conducted using 280 keV ^3He ions to give information on the depth profile of the implanted gallium. The backscattered energy spectra of ^3He incident on the unimplanted area and the region implanted with 10^{17} ions/cm^2 are shown together in Figure 17. Similarly, the unimplanted spectrum is shown with the backscattered spectrum from the 10^{18}/cm^2 -implanted in Figure 18. The depression at the front edge of the spectrum from the implanted regions represents a reduced backscattered yield due to the presence of implanted gallium.

From the kinematic scattering factor for ^3He on zirconium, the surface edge located at channel 724 corresponds to an energy of approximately 246 keV in Figures 17 and 18. The fractional decrease in the two spectra can be used to deduce the concentration of gallium as a function of depth. However, the decreased backscattered spectrum of the implanted area can not be directly subtracted from the unimplanted spectrum to obtain the gallium fraction due to the change in depth scale from the relatively high concentration of gallium (which changes the stopping power of the material).

The TRIM code was used to obtain the stopping power data for ^3He in the Zircaloy that was used for the depth scale calculation. For unimplanted Zircaloy, the stopping power data for zirconium was used. The TRIM code was also used to calculate the stopping power for a range of different Ga-Zr mixtures up to 50% gallium (Ziegler). The stopping power results for the different cases in the energy range of 10-300 keV are shown in Appendix 1. Using the stopping power for zirconium a linear depth scale from the surface energy approximation was estimated at $10.6\text{ }\text{\AA}/\text{keV}$.

To calculate the new depth scale in the implanted regions, the direct difference in the spectra of Figures 17 and 18 was first calculated. This was used as a first

approximation of the gallium fraction in the implanted areas using the estimated depth scale of 10.6 Å/keV. The procedure to calculate the new depth scale was then relatively simple. The small C program shown in Appendix 1 used the numerical method outlined in Chu (Chu, Mayer, and Nicolet, 1978) to calculate the depth scale for the $10^{17}/\text{cm}^2$ and $10^{18}/\text{cm}^2$ spectra. The approach divided the depth of the material into many equal depth slabs of very small thickness. The fractional difference in the spectra from the two implants was used as input to the program. The energy loss of the ^3He ions in the material could then be calculated and related to depth if the stopping power at each location in the material was known and assumed to be constant over the small depth increment. So, to calculate the stopping power at each depth increment, the program used the assumed gallium concentration and interpolated between the calculated stopping power range of Appendix 1. A power function was found to be a very good fit to the stopping power as a function of energy for the TRIM data.

After the new depth scale was calculated, the fraction of gallium was obtained by comparing the backscattered counts from equal depth increments between the implanted and unimplanted areas. Because of the change in depth scale, the calculated Ga concentration increased over the first approximation difference in the spectra because the same depth increment represents more energy channels in the MCA spectrum (Eskildsen, 1981; Smith and Van Wyk, 1974). The result of the calculated depth concentration for the $10^{17}/\text{cm}^2$ implant is shown in Figure 19. Figure 19 shows that the gallium concentration for the $10^{17}/\text{cm}^2$ implant has a roughly gaussian shape that peaks at approximately 350 Å and extends to 650 Å. These depths are in good agreement with the projected range of 409 Å for 100 keV Ga ions in zirconium as calculated from the

TRIM code. Also, the projected range plus one standard deviation in projected range is calculated to be 638 Å, explaining the shape of the curve in Figure 19. Figure 19 also shows that the peak gallium concentration corresponds to approximately 34 % Ga.

Figure 20 shows the gallium profile for the area implanted with 10^{18} ions/ cm^2 . The resulting concentration profile is seen to be relatively flat up to a depth of 500 Å, with the fraction varying from 0.3 to 0.4 gallium present. It is also seen that the gallium extends to a depth of 1150 Å. The significant depth of the gallium indicates that enhanced diffusion is taking place due to the increased disorder and number of vacancies (Eskildsen, 1981). When comparing the concentration of gallium at the surface of Figures 19 and 20, it is seen that the fraction of Ga at the surface increases from 0.08 to 0.26. The fact that this value seems to increase by roughly 3 times and not 10 due to the change in implant fluence can probably be attributed to gallium lost at the surface as a result of sputtering.

It is interesting to note that the concentration of gallium present in the $10^{18}/\text{cm}^2$ area seems to saturate near 40%. The saturation may be explained by the formation of compounds between Ga and Zr with the excess gallium rejected deeper into the Zircaloy. The Ga-Zr phase diagram in Appendix 2, suggests that most likely the compound GaZr_2 would be formed for the concentration of gallium present (Binary Alloy Phase Diagrams, 1990).

Secondary Ion Mass Spectrometry analysis was conducted to supplement the depth profile information from the RBS data. Figures 21-23 show results of SIMS analysis where the beam was positioned at the center of each of the three implanted areas. It should be noted that the concentrations given on the figures are not exact but only approximate, and a transition depth of the first 100 Å is seen on Figures 21-23 where the SIMS results are unreliable. Figure 21 gives the results

from the $10^{16}/\text{cm}^2$ area which shows a typical depth profile shape for ion implantation of Ga in the Zircaloy. The gaussian like distribution

peaks at approximately 400 Å and extends to a depth of 600 Å before dropping an order of magnitude from the peak concentration.

Figure 14: BSE Image and X-Ray Map of 10^{18} Ga/cm² Area

Figure 15: BSE Image of $10^{18}/\text{cm}^2$ Area Showing Position of Line Scan

Figure 16: Electron Microprobe Linescan of $10^{18}/\text{cm}^2$ Area

Figure 17: RBS Spectra of Unimplanted and $10^{17}/\text{cm}^2$ Areas

Figure 18: RBS spectra of Unimplanted and $10^{18}/\text{cm}^2$ Areas

Again, this agrees well with the projected range of Ga of 409 Å.

SIMS data obtained from the $10^{17}/\text{cm}^2$ implant shown in Figure 22 has a similar profile as the $10^{16}/\text{cm}^2$ area, but a peak concentration roughly one order of magnitude above the $10^{16}/\text{cm}^2$ implant as expected. The depth profile shape is also consistent with the RBS analysis of the same area from Figure 19, which shows a peak concentration near 400 Å. It also shows that no significant enhanced diffusion occurs at this fluence. The advantage of SIMS analysis is its relatively high sensitivity. This high sensitivity allows for the detection of gallium to a depth where the concentration is three orders of magnitude lower than the peak concentration, which occurs at 1600 Å for the $10^{17}/\text{cm}^2$ implant.

Finally, Figure 23 shows the SIMS profile from the high fluence implant area of $10^{18}/\text{cm}^2$. Figure 23 suggests that the gallium content has reached a saturation value somewhat greater than the $10^{17}/\text{cm}^2$ implant, which is in agreement with the RBS results. Also in agreement with the RBS results of Figure 20, the SIMS data shows that enhanced diffusion of the gallium has taken place as the bulk of the gallium is now some 500 Å deeper than the $10^{17}/\text{cm}^2$ case. In contrast, the RBS results of Figure 20 shows a relatively flat profile of gallium over the first 600 Å. This may suggest a somewhat greater transition depth of the SIMS analysis. It is further seen that the gallium concentration falls by three orders of magnitude by 2600 Å.

3.4 PHASE IDENTIFICATION

To investigate the presence of intermetallic compound formation between Ga and Zr, an X-ray diffraction spectrum was obtained from the implanted Zirc-4 sample and an unimplanted standard as described in Chapter 2. Figure 24 shows the results obtained from the experiment for the standard. Results from a peak search program along with diffraction data for Zr, Ga, and several Ga-Zr phases and the Ga-Zr phase diagram is located in Appendix 2. Comparing the peak locations to the diffraction data for the α phase of zirconium in Appendix 2, it can be seen that the unimplanted Zirc-4 spectrum is that of α zirconium. This makes sense, when considering Zirc-4 is composed of roughly 98% zirconium.

Figure 25 shows the diffraction spectrum obtained from the Zircaloy sample implanted with gallium. Figure 25 is seen to be virtually identical to the spectrum depicted in Figure 24. Again, the only peaks, which can be identified are due to the zirconium phase, with no new peaks formed as the result of the presence of gallium. Although the RIGAKU diffraction apparatus is capable of detecting phases which may be present in the first 2000 Å of a substrate, the relatively small mass of gallium or Ga-Zr phases which may be present, may be below the detectable limit for the diffraction experiment. It should also be noted that the slit beam could not be completely collimated to impact only the implanted region of the Zirc-4 sample.

Figure 19: Profile of Ga in Zirc-4 Obtained from RBS data for 10^{17} Ga/cm²

Figure 20: Profile of Ga in Zirc-4 Obtained from RBS Data for 10^{18} Ga/cm²

Figure 21: SIMS Results for the Profile of Ga in the $10^{16}/\text{cm}^2$ Area

Figure 22: SIMS Results for the Profile of Ga in the $10^{17}/\text{cm}^2$ Area

Figure 23: SIMS Results for the Profile of Ga in the $10^{18}/\text{cm}^2$ Area

Figure 24: X-ray Diffraction Spectrum from Unimplanted Zircaloy Standard Using Cu K α X-Rays

Figure 25: X-Ray Diffraction Spectrum from Ga Implanted Zircaloy Using Cu K α X-Rays

4. CONCLUSION

The results from the implantation of gallium indicate that the surface of the Zirc-4 is virtually unaffected up to a fluence of 10^{17} Ga ions/cm² which would correspond to a total release of approximately 3 wppm of Ga in a fuel pellet. In addition, no significant pitting or blistering of the surface was observed with a 10^{18} /cm² implant. Sub-grain features on the order of 2 μ m were observed in the surface region of the high fluence, 10^{18} /cm² implant. Electron microprobe measurements indicate that while the gallium concentration is not uniformly distributed in the near-surface region of the Zircaloy, the gallium does not concentrate at grain boundaries.

Depth profile measurements for the region implanted with 10^{18} Ga ions/cm² show that the gallium fraction reaches a saturation value of 0.4 and that significant enhanced diffusion of the gallium takes place far beyond the projected range of Ga ions in Zircaloy. The intermetallic compound GaZr_2 may be formed in the near surface region, which may be present in the form of the observed 2 μ m sub-grains. Such phase formation would help to explain the upper limit that is seen on gallium concentration.

However, x-ray diffraction data failed to identify compound formation.

The results are encouraging for the possible use of WGPu in MOX fuel for the range of Ga that may be present using the dry chemistry fabrication process. Because this study focused on an unstressed Zircaloy sample, the possibility of liquid metal embrittlement cannot be ruled out. However, gallium was not observed to concentrate at grain boundaries. It should also be noted that Ga-Zr compounds are fairly stable over the wide range of temperatures that cladding would experience.

Recommendations for future work include studying the effects of varying the time of ion beam irradiation on Ga diffusion in addition to varying the fluence. Also, to approximate the effects of reactor irradiation, a two step experiment is proposed: introduction of gallium to the near surface-region of Zircaloy followed by heated ion beam irradiation using zirconium or other common fission fragment to simulate fission fragment damage. Transmission electron microscopy (TEM) may prove useful to further characterize phase formation between the Ga and Zr.

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APPENDIX 1 DEPTH SCALE CALCULATION

Calculation of the depth scale for the implanted areas to quantify the gallium profile used the numerical method described in Chu (1978). The method is briefly outlined below along with a copy of the C program used to calculate the depth scale. The procedure began with dividing the depth of the substrate into equal width slabs of width Δx . The energy loss across the n th slab is given by:

$$\Delta E_n = \frac{\Delta x}{\cos \theta} \left. \frac{dE}{dx} \right|_{E_n} \quad (1.1)$$

where θ is the angle between the trajectory of the particle and the surface normal which is equal to 0° on the inward track and 20° along the outward path. The method used in Equation 1.1 assumes that the stopping power is essentially constant.

Stopping powers for 280 keV ^3He were calculated for several different zirconium-gallium mixtures using the TRIM code, the results are shown in Figures 26-31. A power fit was found to be quite close to the data for the stopping powers over the range of 10 - 300 keV. To calculate the depth scale, the gallium content was assumed to be equal to the difference between the two spectrum shown in Figures 17 and 18. Stopping powers needed for the algorithm of 1.1 were based on the gallium content using the surface energy approximation shown in Figures 32 and 33. The stopping power at the different depths was calculated by linearly interpolating between the different Ga-Zr mixtures.

Program Depth.c scans in the assumed gallium concentration as calculated from Figures 32 and 33. It calculates the depth scale using 10 Å depth increments with an incident energy of $E_0 =$

280 keV. The program outputs the depth, backscattered energy, and the stopping power calculated at each depth increment.

Program Depth.c:

```
#include <stdio.h>
#include <stdlib.h>
#include <math.h>

double kinemat(double M, double m_he);
double stop(double E, double diff);
double zr_stop(double E);
double zr_ten(double E);
double zr_twenty(double E);
double zr_thirty(double E);
double zr_forty(double E);
double zr_fifty(double E);

main ()
{
    double SPECT[1601];
    double M = 91.22;
    double m_he = 3.016;
    double k_factor = 0;

    double theta = 0.34907;
    double E = 0;
    double E_o = 280;
    double dx = 0.0001;
    double S_E = 0;
    double E_loss = 0;
    double depth = 0;
    double diff = 0;
    int i = 0;
    int n = 0;
    int k = 1;
```

```

FILE *ifp;
ifp = fopen("thirty", "r");

while (i<1600)
{
    fscanf(ifp, "%lf", &SPECT[i]);
    i=i+1;
}

fclose(ifp);
printf("k    E    depth    S_E\n");
k_factor = kinemat(M,m_he);

while (k < 1500)
{
    n=0;
    E=E_o;

/* Calculation of energy loss proceeding into
the material before scattering. */
    while (n < k)
    {
        diff = SPECT[n];
        S_E = stop(E,diff);
        E_loss = dx*S_E;
        E = E - E_loss;
        n=n+1;
    }

    E = E*k_factor;

/* Calculation of energy loss of exiting
particle after scattering. */
    while (n > 0)
    {
        diff = SPECT[n];
        S_E = stop(E,diff);
        E_loss = S_E*dx/cos(theta);
        E = E - E_loss;
        n=n-1;
    }

    depth = 1*k;
    printf("%d    %f    %f\n",k,E,depth,S_E);
    k=k+1;
}

```

```

}

double kinemat (double M,double m_he)
{
    double p = 0;
    double r = 0;
    double q = 0;
    double K = 0;
    double theta = 2.7925;

    p = sqrt(M*M -
m_he*m_he*(sin(theta)*sin(theta)));
    q = m_he*cos(theta);
    r = m_he + M;

    K = pow(((p+q)/r),2);

    return K;
}

/* Computation of the stopping power at each
depth increment by linear interpolation. */
double stop (double E,double diff)
{
    double S = 0;
    if (diff<=0.0001)
        S = zr_stop(E);
    if (diff>0.0001 && diff<=0.1)
        S = zr_stop(E)-
(diff/0.1)*(zr_stop(E)-zr_ten(E));
    if (diff>0.1 && diff<=0.2)
        S = zr_ten(E)-((diff-
0.1)/0.1)*(zr_ten(E)-zr_twenty(E));
    if (diff>0.2 && diff<=0.3)
        S = zr_twenty(E)-((diff-
0.2)/0.1)*(zr_twenty(E)-zr_thirty(E));
    if (diff>0.3 && diff<=0.4)
        S = zr_thirty(E)-((diff-
0.3)/0.1)*(zr_thirty(E)-zr_forty(E));
    if (diff>0.4 && diff<=0.5)
        S = zr_forty(E)-((diff-
0.4)/0.1)*(zr_forty(E)-zr_fifty(E));

    return S;
}

```

```
/* Stopping power data for Ga-Zr mixtures  
using the fit of Figures 26-31. */
```

```
double zr_stop (double E)
```

```
{  
    double A=0.0262;  
    double B=-0.4629;  
    double stop = 0;  
    stop = 1/(A*pow(E,B));  
    return stop;  
}
```

```
double zr_ten (double E)
```

```
{  
    double A=0.0275;  
    double B=-0.4669;  
    double stop = 0;  
    stop = 1/(A*pow(E,B));  
    return stop;  
}
```

```
double zr_twenty (double E)
```

```
{  
    double A=0.0288;  
    double B=-0.4704;  
    double stop=0;  
    stop = 1/(A*pow(E,B));  
    return stop;  
}
```

```
double zr_thirty (double E)
```

```
{  
    double A=0.0303;  
    double B=-0.4748;  
    double stop=0;  
    stop = 1/(A*pow(E,B));  
    return stop;  
}
```

```
double zr_forty (double E)
```

```
{  
    double A=0.0321;  
    double B=-0.4796;  
    double stop=0;  
    stop = 1/(A*pow(E,B));  
    return stop;  
}
```

```
double zr_fifty (double E)
```

```
{  
    double A=0.0341;  
    double B=-0.4849;  
    double stop=0;  
    stop = 1/(A*pow(E,B));  
    return stop;  
}
```

Figure 26: Stopping Power Data for ^3He in Zr from TRIM

Figure 27: Stopping Power Data for ^3He in 10% Ga - Zr from TRIM

Figure 28: Stopping Power Data for ^3He in 20% Ga - Zr from TRIM

Figure 29: Stopping Power Data for ^3He in 30% Ga - Zr from TRIM

Figure 30: Stopping Power Data for ^3He in 40% Ga - Zr from TRIM

Figure 31: Stopping Power Data for ^3He in 50% Ga - Zr from TRIM

Figure 32: Linear Depth Profile of assumed Ga Concentration for $10^{17}/\text{cm}^2$ Implant

Figure 33: Linear Depth Profile of Assumed Ga Concentration for $10^{18}/\text{cm}^2$ Implant

APPENDIX 2

X-RAY DIFFRACTION DATA AND GA-ZR PHASE DIAGRAM

This section contains diffraction data for Zr as well as Ga-Zr compounds obtained from the International Center for Diffraction Data. The data shown gives the d- spacing and relative intensities of diffracted x-rays for Cu K α x-rays. Following the diffraction data,

output from a peak search program is displayed for the spectra of Figures 24 and 25. Appendix 2 also shows the Ga-Zr phase diagram in Figure 34 (Binary Alloy Phase Diagrams, 1990).

Diffraction Data

Material:	d(Å)	Int.	h	k	l
Zirconium (Zr)	2.798	33	1	0	0
System: Hexagonal	2.573	32	0	0	2
a: 3.232 c: 5.147	2.459	100	1	0	1
Wavelength = 1.54184	1.894	17	1	0	2
	1.616	17	1	1	0
	1.463	18	1	0	3
	1.399	3	2	0	0
	1.368	18	1	1	2
	1.350	12	2	0	1
	1.287	4	0	0	4
	1.2296	4	2	0	2
	1.1689	3	1	0	4
	1.0842	4	2	0	3
	1.0588	2	2	1	0

Material:	d(Å)	Int.	h	k	l
Gallium Zirconium (GaZr ₂)	2.72	8	0	0	2
System: Tetragonal	2.63	100	2	2	1
a: 6.708 c: 5.437	2.37	45	2	2	0
	2.12	55	3	1	0
	1.787	1	2	2	2
	1.758	2	3	2	1
	1.676	3	4	0	0
	1.582	3	3	3	0
	1.559	10	4	1	1
	1.550	19	2	1	3
	1.428	13	4	0	2
	1.367	16	3	3	2
	1.214	16	5	1	2
	1.186	4	4	4	0
	1.144	9	3	1	4

Material:	d(Å)	Int.	h	k	l
Gallium Zirconium (Ga ₂ Zr ₃)	3.45	3	2	1	0
System: Tetragonal	3.18	6	1	1	1
a: 7.718 c: 3.909	2.746	60	2	0	1
	2.587	100	2	1	1
	2.441	50	3	1	0
	2.141	12	3	2	0

2.070	6	3	1	1
1.955	18	0	0	2
1.930	4	4	0	0
1.878	16	3	2	1
1.819	3	3	3	0
1.701	12	2	1	2
1.649	9	3	3	1
1.526	20	3	1	2

The results from the peak finding program in the x-ray laboratory for the unimplanted standard and implanted Zircaloy

samples (see Figures 24 and 25) are shown in Tables 3 and 4 below.

Table 4: X-Ray Peak Intensities and Locations for the Unimplanted Standard

Peak #	d-space	2-theta	I/I _{max}
2.7615		31.986	6.7
2.5811		34.639	100.0
2.4634		36.385	47.9
1.8973		47.557	30.1
1.6184		56.581	3.2
1.4646		63.481	60.5
1.3690		68.493	9.2
1.2881		73.420	11.6
1.1703		82.343	13.7
1.0848		90.530	5.1
1.0071		99.849	15.1

Table 5: X-Ray Peak Intensities and Locations for the Implanted Sample

Peak #	d-space	2-theta	I/I _{max}
2.8041		31.980	5.9
2.5748		34.815	100.0
2.4598		36.498	58.7
1.8968		47.920	26.6
1.6155		56.954	3.8
1.4642		63.483	50.7
1.3689		68.490	8.7
1.2879		73.427	10.9
1.1696		82.385	10.8
1.0843		90.534	5.5
1.0355		96.131	2.6
1.0067		99.841	13.3

Figure 34: Ga - Zr Phase Diagram