AMM Newsletter
Advanced Methods for Manufacturing
Issue 4 • September 2016

Development of nuclear quality components using metal additive manufacturing

radiabeam SYSTEMS

P. Frigola, RadiaBeam Systems, LLC

A. Hinojos, J. Mireles, I. Segura, L. E. Murr, R. B. Wicker, W. M. Keck, Center for 3D Innovation, University of Texas at El Paso

Peter Hosemann, A. Reichardt, Department of Nuclear Engineering, University of California at Berkeley

Multi-material parts enhance functionality and performance in a variety of applications in the nuclear power industry. A major challenge in manufacturing multi-material components is the joining of dissimilar metals. Traditional joining methods, including brazing, welding, and soldering, can result in the formation of precipitates, intermetallics, and distortions at the weld interface that are detrimental to the part's performance.

This project aims to develop, optimize, and commercialize the electron beam melting (EBM) additive manufacturing (AM) process for joining austenitic steels to nickel-based superalloys, as well as develop the capability (using EBM AM) to efficiently join ferritic to austenitic steels components for use in the nuclear power industry.

EBM AM is a fabrication process that uses a focused electron beam to fully melt metal powder in a layer-by-layer fashion. The use of an electron beam makes the energy deposition process very efficient, fully melting a variety of metallic powders in an evacuated processing environment, resulting in limited contamination of oxides and nitrides, and providing a high-quality metallurgical joint while minimizing the thermal damage to surrounding material.

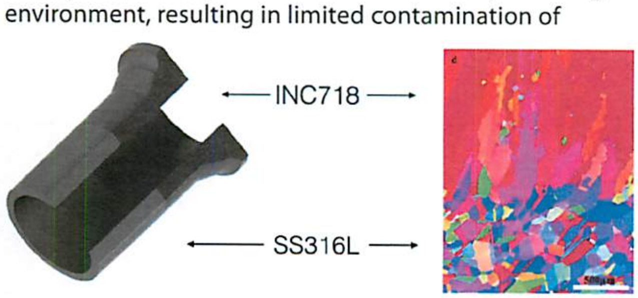

Figure 1. Concept illustrating dissimilar metal joining using additive manufacturing fabrication (left), and electron backscatter diffraction image of electron beam melting additively manufactured INC718 on SS316L (right).

This multi-disciplinary collaborative project, comprised of RadiaBeam Systems, the University of Texas at El Paso W.M. Keck Center for 3D Innovation, and the University of California at Berkeley, has focused on the development of EBM AM process parameters and technology to join Inconel 718 (INC718) and Inconel 690 (INC690) alloys to 316L stainless steel (SS316L). Figure 1 illustrates the concept of multi-material AM, along with electron backscatter diffraction (EBSD) image of EBM AMed INC718 on SS316L.

Continued on next page

In This Issue

For more program information, including recent publications, please visit www.energy.gov/ne

Continued from previous page

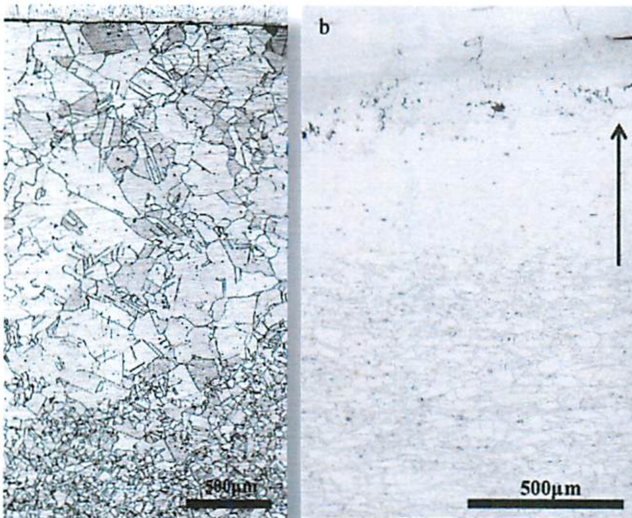

Figure 2. Micrograph reconstruction revealing the heat affected zone of the electron beam melting processed SS316L (left) and INC718 (right) substrates; the arrow indicates electron beam melting additive manufacturing build direction. Analysis reveals finer weld joints when compared to traditional welding methods.

Figure 3. Inverse pole figure (IPF) color map derived by electron backscatter diffraction of electron beam melting fabricated INC718 on SS316L (left), and SS316L on INC718 (right). Micrographs show columnar grains with a preferred texture growing from substrate grains. The dotted lines depict area of fusion.

Current Status

This project has experimentally demonstrated the feasibility of joining INC718 and INC690 with SS316 using EBM additive manufacturing. Multi-material components have been fabricated using EBM AM, and the joint interface characterized.

Characterization of the EBM INC718 on SS316L interface reveals minimal thermal effects (e.g., reduced presence of precipitates) and heat affected zone (HAZ) depths as small as 443±56µm (see Figure 2). Results of the INC718-SS316L EBM fabrication have been recently published (*A. Hinojos, et. al., Material & Design, Vol. 94, 15 March 2016, pages 17–27*).

As noted by A. Hinojos et. al., the columnar growth inherent in the EBM fabrication process has the potential for part repair and feature addition with a tailored microstructure that joins epitaxially from the substrate. Figure 3 depicts columnar grains with a preferred texture growing from the substrate grains.

In the past 6 months, EBM AM process parameters have been developed for joining of INC690 to SS316L, and samples suitable for metallographic and mechanical characterization have been fabricated. Detailed microstructural and mechanical analysis is currently ongoing. Figure 4 shows preliminary mechanical properties of the newly processed INC690.

Irradiation parameters and a target design (see Figure 5) have also been developed during this time. Initial irradiation testing of INC718/316LSS and INC690/316LSS targets will be carried out at LANL IBML. The target will be heated 320°C to simulate light water reactor operating temperature, and monitored via several thermocouples. The samples will be exposed to a 1.5 MeV proton beam at ~10 µAmps (for a dose of ½ to 1 DPA), penetrating the material to a depth of 15–20 µm.

Conclusion

EBM AM continues to show excellent promise for the efficient (cost-effective) production of multi-material parts for the nuclear power industry. The feasibility of joining INC718 and INC690 to SS316L has been established, and the EBM AM process has been shown to produce parts with improved joint qualities compared to traditional welding methods. Further EBM parameter optimizations for specific geometries of interest, along with extensive characterization, are planned for the near future.

Continued on next page

Continued from previous page

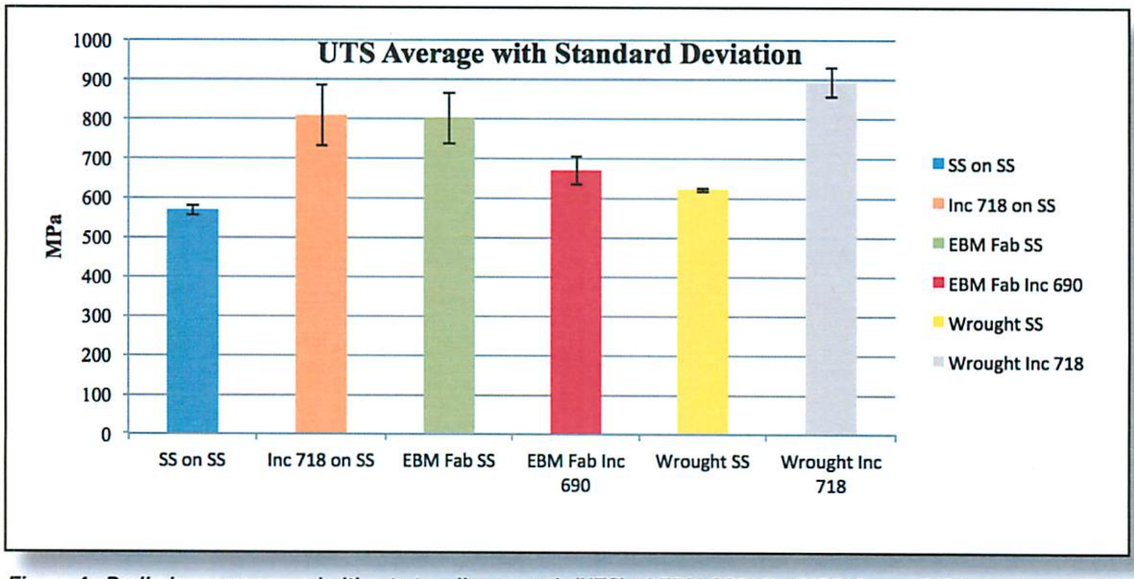

***Figure 4.** Preliminary measured ultimate tensile strength (UTS) of EBM SS316L, INC718, and INC690 compared to measured UTS for wrought SS316L and INC718.*

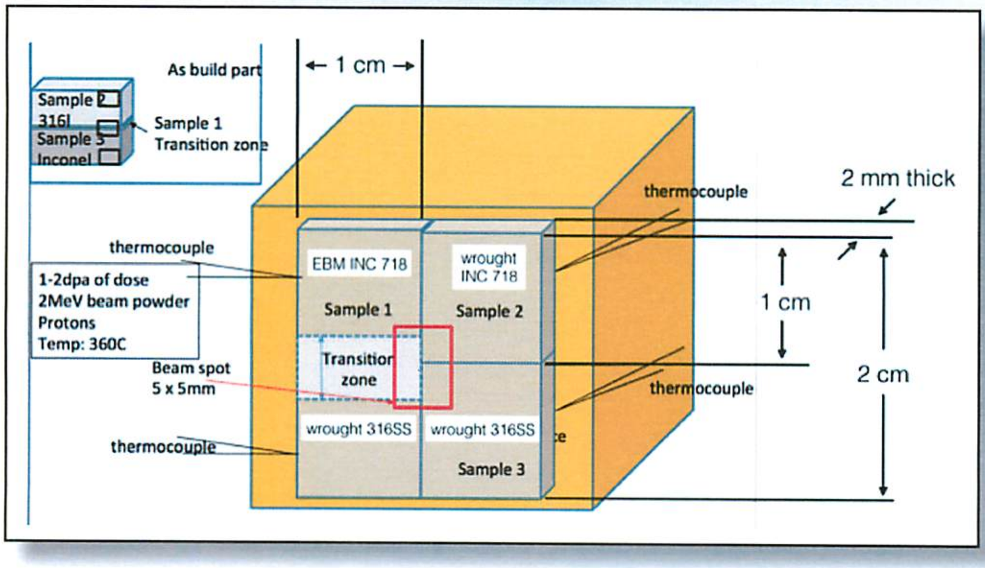

***Figure 5.** Irradiation target design.*

Improving Weld Productivity and Quality by means of Intelligent Real-Time Close-Looped Adaptive Welding Process Control through Integrated Optical Sensors

Jian Chen (PI), Roger Miller, Zhili Feng, Oak Ridge National Laboratory (ORNL), Oak Ridge, TN 37831
Yu-Ming Zhang, University of Kentucky, Lexington, KY 40506
Robert Dana Couch, Electric Power Research Institute (EPRI), Chattanooga, TN 37405

Jian Chen
Oak Ridge National Laboratory

Welding is one of the most important manufacturing technologies in fabricating nuclear reactors. Eliminating weld defects is crucial due to the detrimental effects on the component integrity and safety. Today, welds are made in a prescribed manner. Welding conditions, such as welding current and welding speed, are pre-determined based on weld qualification trials. It is very difficult, if not impossible, to proactively adjust in real-time the welding conditions to compensate for unexpected variations in real-world welding that cause the formation of welding defects. Post-weld quality inspections such as ultrasound, x-ray, and dye-penetrant are generally mandatory per code requirement. Repair and correction of the weld defects after the weld is made is time-consuming and expensive—often much more than the cost of initial welding. Furthermore, undetected weld defects left in the structure are a major threat to the safety and structural integrity of nuclear reactor structural components. In this 3-year program, Oak Ridge National Laboratory (ORNL) is collaborating with University of Kentucky and Electric Power Research Institute (EPRI) to develop a novel close-looped adaptive welding quality control system based upon multiple optical sensors. It will enable real-time weld defect detection and adaptive adjustment to the welding process conditions to eliminate or minimize the formation of major weld defects typically encountered when welding high-performance engineering structural materials for nuclear structural components. The multi-optical sensing system (Fig. 1) consists of a visible (VIS) camera, an infrared (IR) camera, a weld pool surface measurement sensor, and the necessary auxiliary illumination sources and filters. The sensing system will be capable of simultaneously measuring, in real-time, the changes in the welding temperature field via IR camera, the strain and stress fields based on digital image correlation (DIC) via VIS camera, and the dynamic changes of weld pool surface. The measurement signals will be correlated to the weld quality and provide feedback control of the welding processes.

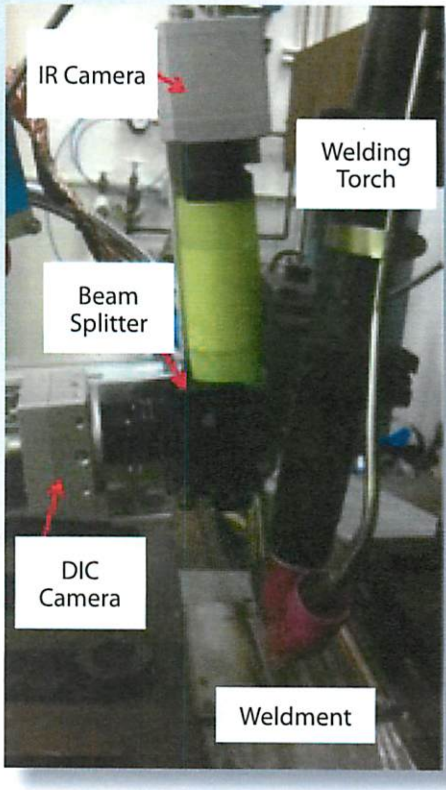

Figure 1. Multi-optical sensing system

Impact and value to nuclear applications

In today's industry, the weld quality inspection for manufacturing nuclear reactor structures mostly relies on post-weld non-destructive examination (NDE) techniques. If a defect is identified, the reworking (or scrapping if beyond repair) of manufactured structures are costly and time-consuming, particularly for the thick-section reactor structures. The multi-sensing system monitors and controls the weld quality in real time. By drastically reducing weld defects, and therefore the rework required for defect mitigation, the online system can significantly decrease the component fabrication cost, accelerate the deployment schedule, and increase the integrity and reliability in a variety of nuclear reactor designs and components.

Continued on next page

Continued from previous page

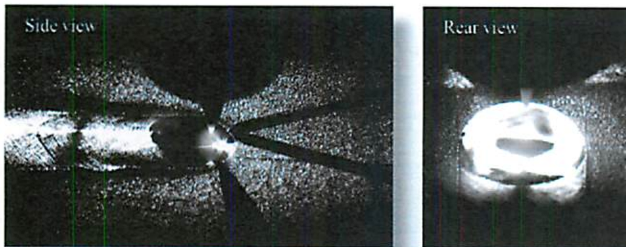

Figure 2. *Arc Weld pool visualization during gas tungsten arc welding by a special illumination and optical filtering system*

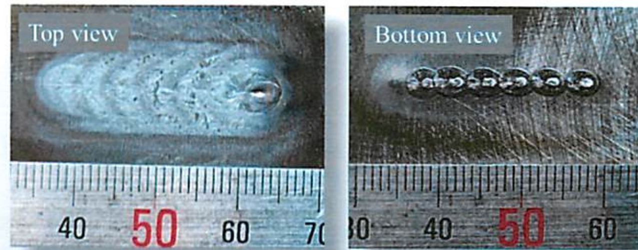

Figure 4. *Full penetration weld bead of a Gas tungsten arc welding process was achieved*

Current Status

The multi-sensing system, including hardware and software, has been integrated and tested in various welding conditions. The major achievements since the beginning of the project include.

- Applying a special auxiliary illumination and optical filtering technique to suppress the intense arc light effectively and visualize the liquid pool and the surrounding area (Fig. 2).
- Developing a unique real-time strain and stress monitoring method based on DIC. First, the aforementioned auxiliary optical system has provided a stable illumination source that is insensitive to the intense welding arc. Second, a high-temperature surface speckle patterning method has been developed. The speckle pattern, required by the DIC algorithm, survives at the elevated temperature up to the metal's melting point. Third, a special algorithm has been developed to determine, in real time, the evolution of both strain and stress adjacent to the fusion line from the DIC and the temperature measurements (Fig. 3).
- Achieving real-time monitoring and control of weld penetration using a special weld pool surface measurement sensor (Fig. 4).

Conclusion

In FY 2016, the multi-optical sensing system has been integrated and tested in a variety of welding conditions. Efforts have been made successfully to measure, in real time, the strain and stress states adjacent to the fusion line, and the dynamic changes of the weld pool surface. The signatures of the multi-optical signals have been analyzed and compared to the weld characteristics to establish the database that will be further utilized in adaptive process control. The preliminary results have demonstrated the feasibility of full-penetration control during the Gas tungsten arc welding (GTAW) processes.

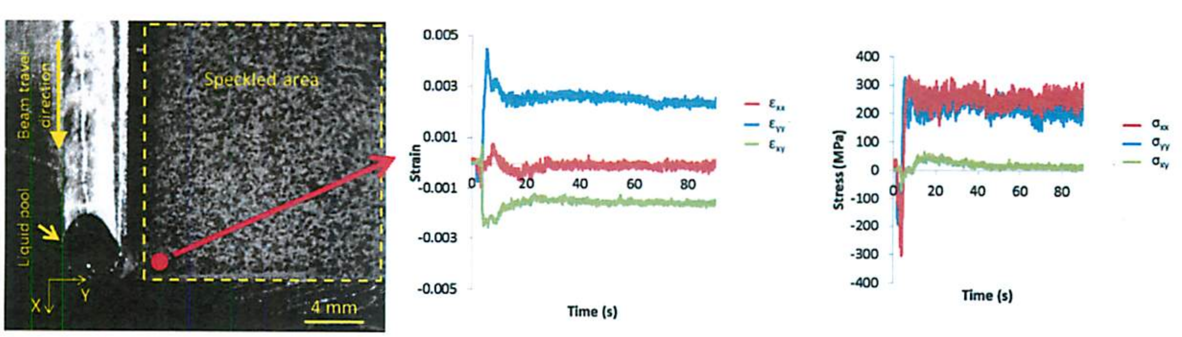

Figure 3. *Real-time strain and stress monitoring during a laser welding process*

Advanced surface plasma nitriding for development of corrosion resistant and accident tolerant fuel cladding

Lin Shao and Frank Garner, Texas A&M University, College Station, TX 77845
Don A. Lucca, Oklahoma State University, Stillwater, OK 74078
Michael Short, Massachusetts Institute of Technology, Cambridge, MA 02139




Although various surface coating techniques have been proposed to increase oxidation and corrosion resistance of fuel cladding materials, the de-bonding of the coating layer with the original cladding matrix under exposure to coolants makes such approaches unsuitable for reactor applications. Furthermore, the feasibility of techniques for large-scale processing on cladding tubes remains another technological bottleneck. This project aims to develop a hollow cathode plasma nitriding technique to solve the above issues. Starting with alloys of interest to the nuclear energy community, the team will apply an advanced surface plasma nitriding technique to convert alloy surface layers into nitride layers for better structural integrity and compatibility with both coolants and nuclear fuels. The technique can form uniform nitride layers on both inner and outer surfaces of cylindrical hollow tubes. The treated and untreated samples will be irradiated by using Fe self-ions or dual beams of Fe ions and He ions, up to extreme damage levels (>400 dpa) and at various temperatures (300°C to 800°C). Various microstructural characterizations and mechanical property measurements will be performed. Samples will be tested in high-temperature water and liquid sodium coolant loops to evaluate their corrosion resistance and compatibility with coolants for light-water reactor (LWR) and fast reactor applications.

Impact and Value to Nuclear Applications

In reactor harsh environments involving high-stress, high-corrosion and high-irradiation damage, fuel cladding materials experience severe structural changes and degradation, which impacts not only lifespan but also reactor safety and reliability. Development of advanced fuel cladding materials is critical for both present designs with extended life times and advanced designs with even harsher operation conditions. Although numerous studies have shown that a surface nitride layer can be used to increase hardness, wear resistance, oxidation resistance, and corrosion resistance, a systematic investigation has not been performed pertaining to nitriding effects on fuel cladding materials, particularly when neutron damage and fuel cladding interactions must be considered. Furthermore, existing plasma nitriding techniques are not able to uniformly modify a hollow tube.

The importance and relevance of the project to the Office of Nuclear Energy (NE) program is reflected by the following benefits.

- The development of a new plasma nitridation technique, which is able to uniformly nitride fuel cladding tubes, including both the outer and inner tube surface.
- The starting materials are of great interest to the Department of Energy Office of Nuclear Energy (DOE-NE) mission. Grade 92 and Alloy 709 have been targeted for fast reactor applications because of their high-temperature stability and mechanical properties. EK 181 belongs to the low-activation ferritic/martensitic alloys that have reduced radioactivity during spent fuel storage or a core accident. Zircaloy 2 and 4 are cladding materials being used for the present LWR design and their behavior for extended lifetimes needs to be investigated.
- The cladding is virtually immersed in an N plasma, and N atom bombardments come from all directions, which make it feasible to treat materials of arbitrary shapes (i.e., cladding tubes having internal and external surfaces). This represents a great advantage when compared with traditional ion implantation.

Continued from previous page

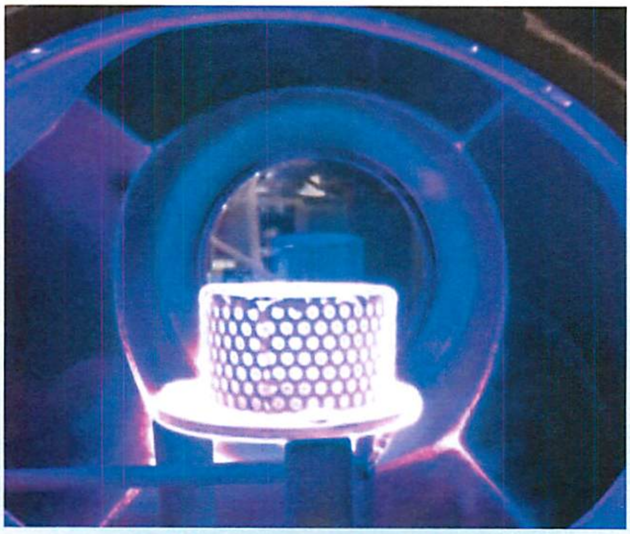

Figure 1. Image of hollow cathode plasma chamber, with Ar plasma created under AC superimposed on 900V DC. The device was built in the PI's lab.

- The nitride films or nitride particles form inside the original cladding matrix, which guarantees structural integrity. This represents a great advantage when compared with other techniques, such as physical vapor deposition or laser ablation.
- Mechanical properties, such as wear resistance and hardness of the cladding surface, can be significantly enhanced due to nitride dispersion.
- The nitride films or particles can introduce compressive stress at the surface and greatly enhance the critical stress required for crack initiation. This may further impact the resistance for delayed hydride cracking during dry storage.
- Plasma nitriding can be performed over a broad temperature range. Therefore, steels can be nitrided without forming chromium nitride precipitates, which maintains the steels' corrosion resistance.
- Nitriding can be performed at very low temperatures that are significantly less than the last tempering temperature of the cladding materials, which sustains the internal microstructure required for optimized mechanical properties.
- The surface nitride, either as particles or as a continuous film, is expected to enhance radiation tolerance of the cladding due to introduced boundaries, which act as defect sinks.
- The plasma-treated cladding materials do not need additional machining or polishing. They are ready to use after the process.

Current Status

Different from the conventional plasma nitriding process, the technique of Cathodic Cage Plasma Nitriding (CCPN) was employed. As shown in Fig. 1, a nitriding cage is introduced to create the hollow cathode effects that result in an optimal pressure depending on the size of the holes of the cage. In comparison with the traditional approach, the CCPN technique minimizes edge effects, increases temperature uniformity, and reduces arcing. Thus, the design is more suitable for samples having complex geometry. The three most important variables in the nitriding process are pressure, voltage, and time.

Continued on next page

Continued from previous page

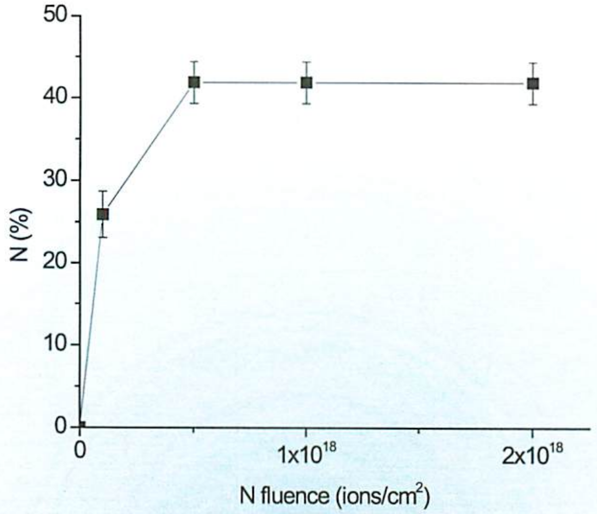

Figure 2. N concentrations (atomic percentage) in the N peak region as a function of N fluences in pure Fe.

After optimization of the plasma chamber, systematic nitriding experiments were performed on four sets of samples: pure Fe, pure Zr, zircaloy-4, and HT-9. The pressure is controlled to be 1 Torr and nitriding time is fixed to be 4 hours. The nitriding temperatures are 350°C, 400°C, 450°C, and 500°C, respectively. For a comparison study, samples were also irradiated by nitrogen ions using accelerators to study phase evolution under precisely determined nitrogen fluences. After nitriding, samples are characterized by Rutherford backscattering spectrometry (RBS) to obtain composition information.

Atomic scale characterization was used to determine the morphology and phases of nitride layers forming after the plasma treatment. As one example, Fig. 2 shows that under nitrogen ion bombardment, surface layers of pure Fe begin to evolve into a nitrogen-rich layer with increasing nitrogen concentrations. Upon a critical nitrogen fluence, a nitride layer forms with saturated nitrogen concentrations, suggesting formation of a stable nitride layer. Fig. 3 shows transmission electron microscopy image, localized diffraction pattern, and fast Fourier transform (FFT) pattern, which are used to determine nitride phase in pure Fe as a model material for stainless steels. In addition to Fe_3N, another nitride phase was identified that agrees with theoretically predicted FeN_2 (R3m crystal structure), but it was never reported before from experimental studies.

By controlling nitridation time, the structures change from precipitation-like nitride, which are embedded in original matrix to continuous nitride layers. Another key interest is to study thermal stability of nitride layers. All nitride samples were thermally annealed up to 600°C for various annealing times, with in situ RBS performed to obtain kinetics of nitride dissociation, to identify the operation limits of these materials. Mechanical property measurements and structural integrity tests under harsh coolant conditions are currently in progress.

Conclusion

The first-year study demonstrates the feasibility of the technique to form nitride layers on various alloys relevant to the nuclear energy community. The chamber design was optimized to achieve the best nitridation performance. The key parameters that influence morphology and phases of the nitride layers have been identified. The project has gained the knowledge on thermal stability of various nitride layers and performance limitations of these surface-treated materials at elevated temperatures. Currently, radiation responses, corrosion resistance, and mechanical property changes of plasma treated alloys are being studied for the next phase of this analysis.

Continued on next page

Continued from previous page

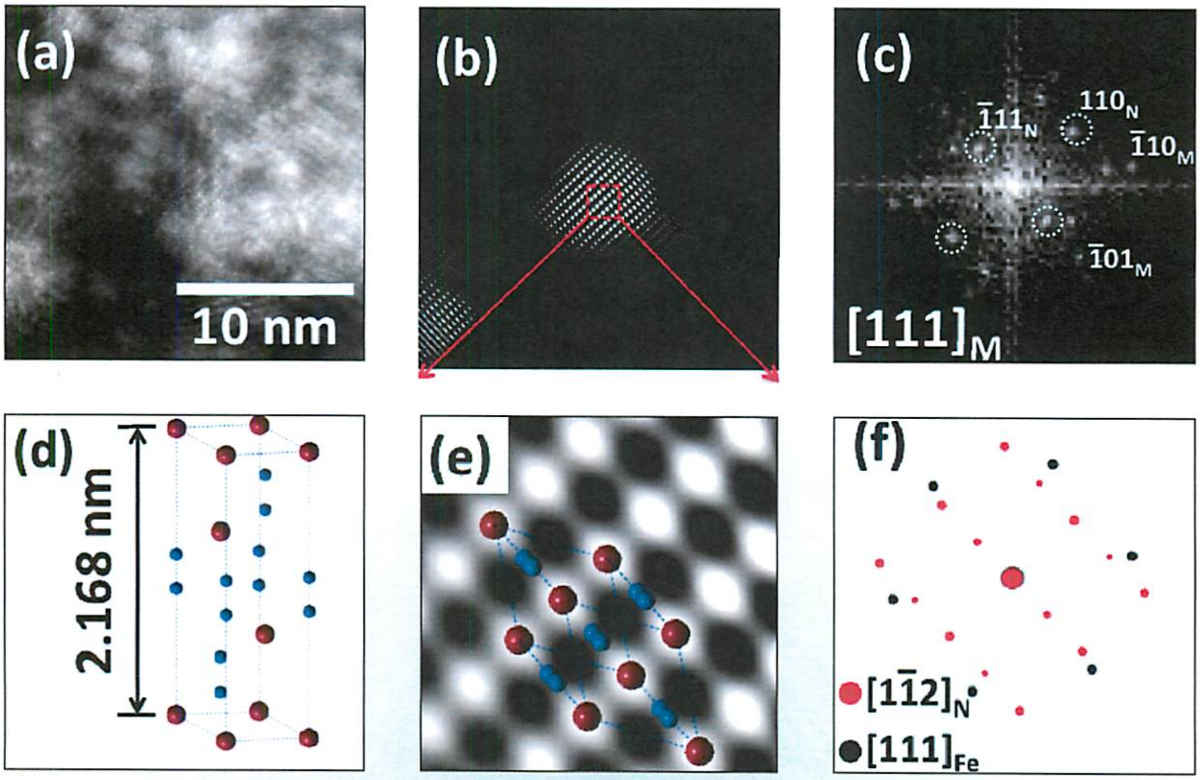

Figure 3. *(a) a high-resolution cross sectional TEM view of N-implanted Fe, and (b) the corresponding inversed FFT pattern and (c) the corresponding indexed FFT pattern with circled spots used to construct (b), and (d) schematics of FeN_2 crystal structure with blue color referring to N atoms and red color referring to Fe atoms, and (e) the comparison of inverse FFT pattern and projected view of FeN_2 crystal structure, and (f) simulated diffraction patterns.*

To submit information or suggestions, contact Alison Hahn at **Alison.Hahn@nuclear.energy.gov**.

15-GA50081-04-R4