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HIGH-RATE RADIATION DAMAGE STUDIES OF MATERIALS WITH HEAVY ION BEAMS*

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Abstract

The Argonne Tandem Linear Accelerator System (ATLAS) at Argonne National Laboratory is a superconducting ion linac capable of delivering beams ranging over all possible elements, from hydrogen to uranium, and at a wide range of beam currents and energies. The ATLAS scientific program is focused primarily on basic nuclear physics. In this contribution, we present the capabilities of ATLAS for high-rate radiation-damage studies for a variety of applications below the threshold of producing radioactivity. To date ATLAS has been used for such studies relevant to advanced nuclear reactors. These include studies of radiation damage in structural materials and damage induced by fission products in advanced fuel candidates. Such studies can be expanded to include in-situ measurements of response to radiation damage in other materials used at high power densities such as for targets at spallation neutron sources and neutrino factories. ATLAS is in the process of a multi-user upgrade which adds the capability of simultaneously accelerating two ion beams and delivering them to different target stations. This enables ATLAS to deliver beams for nuclear physics research simultaneously with materials irradiation studies.

INTRODUCTION

ATLAS Capabilities

ATLAS can provide beams ranging across the periodic table, from hydrogen to uranium [1]. Figure 1 demonstrates the versatility of ATLAS, showing 38 unique ion beam species delivered in 2018. Three acceleration stages provide energies ranging from a few hundred keV per nucleon up to 20 MeV per nucleon, depending on the specific ion and beamline [2]. The intensities of these beams range from as low as a few particles per second up to 10 microamps.

Previous irradiation damage studies in nuclear reactor fuel and structural materials at ATLAS have demonstrated the ability to utilize the facility for this type of studies [3]. Those studies paved the way for the recent developments; ATLAS recently collaborated with Argonne's Nuclear Science and Engineering and Chemical and Fuel Cycle Technologies divisions to commission the ATLAS Material Irradiation Station (AMIS) [4,5]. AMIS is a new beamline and experimental station located downstream of the first stage of acceleration and 100 degrees off-axis from the zero-degree beamline (Fig. 2). This new beamline is dedicated solely to ion beam irradiation studies of materials. In the next section of this paper, we will discuss nuclear fuel and structural materials studies currently being performed at AMIS by Argonne's Nuclear Science and Engineering and Chemical and Fuel Cycle Technologies division.

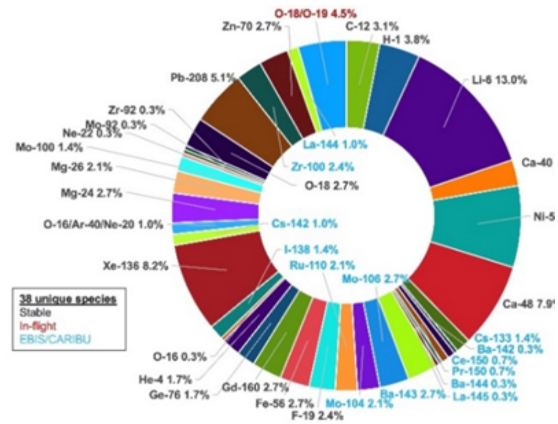

Figure 1: 2018 ATLAS beam species.

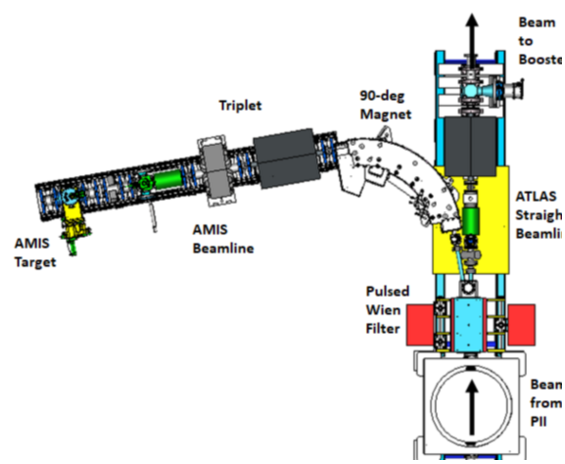

Figure 2: The new AMIS beamline and ATLAS zero-degree line.

In addition to AMIS, ATLAS has available beamlines after the second and third stages of acceleration which could be used for a wide range of ion beam irradiation applications. We will also discuss in the next section the feasibility of using available ATLAS ion species and energies to perform radiation damage studies on candidate materials for applications in high-power targets.

RADIATION DAMAGE STUDIES

Nuclear Fuel and Structural Materials

Nuclear fuel experiences a uniquely harsh environment during its life in a reactor. Maintaining its structure under transmutation as well as lattice damage from neutrons and fission fragments, is key to fuel lifetime for many reactor types. However, determining the fuel's response to the

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many different mechanisms is typically only done by in-reactor testing. While this type of test is ultimately necessary to confirm fuel performance, in-reactor testing is lengthy, expensive, and involves many testing conditions and combined effects. Heavy ion irradiation of nuclear fuel can be used as a screening experiment to better utilize in-reactor tests, as well as a method to isolate and understand specific phenomena, the results of which can be fed into fuel performance modelling. The AMIS beamline is used to examine fission gas bubble evolution, growth of interaction layers between fuel and cladding or surrounding matrix materials (in dispersion fuel), and ballistic collisions cascade damage in structural materials. A simplified representation of these phenomena is shown in Figure 3.

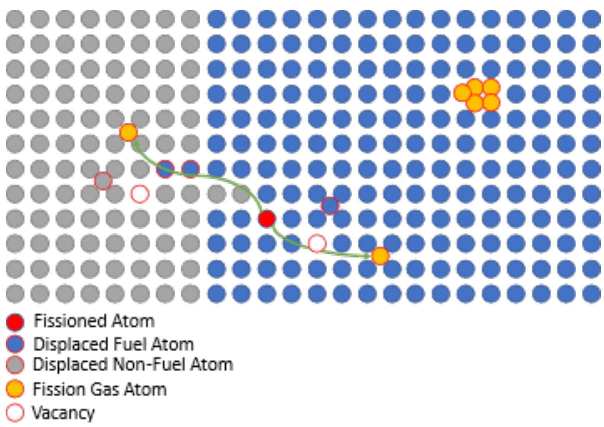

Figure 3: A simplified diagram of some of the modifications to fuel and nearby structural material due to fission events that are isolated and studied by ATLAS irradiations. Mixing of interfaces, the formation of interstitial and vacancy defects, as well as the accumulation of gaseous fission products into bubbles are represented.

In the bulk of the fuel, a gas atom produced by fission travels a short distance and then stays trapped in the lattice. As the density of gas in the fuel increases, it tends to form bubbles at various lattice sites. The geometric swelling and decrease in thermal conductivity of the fuel from fission gas bubble growth is detrimental to fuel lifetime. Therefore, it is necessary to understand their temperature, morphology, and gas density dependence. Historically, the simulation of fission gas bubble growth through the implantation of 84MeV Xe has been studied [6-9] at ATLAS. Experiments are planned to coordinate reactor and ion irradiations of metallic and ceramic fuels to directly compare fission gas bubble growth, enabling the much higher gas densities that AMIS can achieve to be used to guide reactor irradiations.

In dispersion fuel systems, interaction occurs between the fuel particles and the surrounding matrix material, which accelerates mixing from fission fragments. The formation of an interaction layer in the fuel system, if extensive, typically leads to adverse effects [10]. Fully decoupling the temperature and dose effects on interaction layer growth is necessary for modeling but is difficult for in-reactor testing. Heavy ion irradiation studies have successfully probed this phenomenon [11] and are actively planned for various fuel systems.

The range of heavy ions at these energies, which allow for interaction layer studies, also provides an improved geometry for studying coatings and structural materials. The response of structural materials to knock-on damage caused by neutrons is similarly difficult due to the activation of the material by neutron irradiation on top of the challenges of reactor irradiations. Lower energy ion irradiations have been frequently used to better understand the response of materials to damage cascades. However, their range and, therefore, volume of material affected is small, limiting the types of analysis [12]. AMIS can provide a multi-micron deep irradiated region free from surface effects and implanted region in typical structural materials to study the stability of thicker coatings and damage evolution in nuclear structural materials [13, 14].

High Power Targets

Targets capable of handling high-power beams are crucial for many fields of study, including the production of neutrinos and spallation neutrons. Over time changes to the lattice of the target due to radiation damage will change its material properties, eventually leading to its failure. The target survivability and lifetime have been the limiting factors to the beam power that can be delivered on target rather than the accelerator itself [15]. R&D into novel high-power targets is essential for existing and future multi-megawatt facilities to achieve maximum beam power on targets [15,16].

Displacement per atom (DPA) is the commonly used unit to measure radiation damage to targets and beam intercepting devices, as well as a measure to understand the effects of this damage on a micro- and macroscopic scale [17]. Although high-power accelerator target facilities often use intense and energetic beams of protons; similar radiation damage as measured in DPA's can be achieved at a lower energy facility like ATLAS, using higher mass beams.

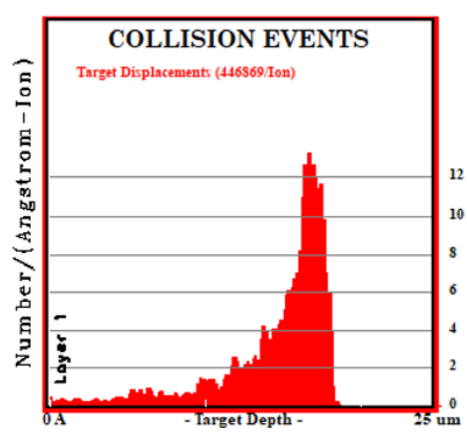

Figure 4: Total target displacements from a 5 MeV/u W beam on a 25 µm thick W foil from SRIM.

Tungsten (W) and W alloys are popular materials for high-power targets because of their high density and melting point [15]. ATLAS can deliver a 5 MeV/u beam of W

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onto a thin W foil located after the third stage of acceleration. Self-ion irradiation prevents the accumulation of impurities which can affect the properties of interest. The Monte Carlo simulation code SRIM [18] predicts ~400000 displacements of W atoms per W ion stopping in 25 microns of W foil (Fig.4), from this one could then calculate the number of DPA's. In-situ measurements, such as electrical conductivity vs. DPA's could then be made on the W foil while being irradiated.

MULTI-USER UPGRADE

ATLAS is the only DOE low-energy stable beam nuclear physics user facility. As such, there is enormous competition for beamtime and an increasing demand for more extended experiments (>1 week). ATLAS is in the process of being upgraded for multi-user operations to meet this demand and broaden the potential user community.

The low-energy beam transport will be modified to support the simultaneous injection and acceleration of two beams with very close mass-to-charge ratios: one stable from an Electron Cyclotron Resonance (ECR) ion source and one radioactive from CARIBU electron beam ion source (EBIS) charge breeder CARIBU-EBIS. CARIBU stands for "Californium Rare Isotope Beam Upgrade". The planned beam composition is 3% for the radioactive beam, 96% for the stable beam, and 1% required for switching [4].

The stable beam will soon be able to be extracted at the AMIS beamline via a Wein filter magnet (Fig 2) for pulsed switching between CARIBU-EBIS and ECR beams. This pulsed beam extraction after PII would allow for heavy-ion beams of 1–1.5 MeV/u to be delivered to the AMIS beam line for irradiation studies while CARIBU-EBIS beams are simultaneously transported into the following stages of acceleration and delivered to downstream end stations for nuclear structure studies.

A second switch yard is being designed to allow for beam extraction after the second stage of acceleration, with the stable portion of the beam being able to be delivered to an irradiation station at 5-7 MeV/u and the radioactive portion of the beam moving along the beamline to the third stage of acceleration.

CONCLUSION

In conclusion, the ATLAS particle accelerator at Argonne National Laboratory provides an ideal platform for radiation damage studies due to its wide range of capabilities, including its ability to produce a variety of ion species at different energies. A successful study of radiation damage to nuclear fuels is ongoing at ATLAS. Additionally, the heavy ion beams available at ATLAS could enable researchers to simulate radiation damage that materials and components would receive from highly energetic beams of protons without producing radioactivity. Radiation damage studies could take place now after each of three stages of acceleration. In addition, once the multi-user upgrade is completed, additional beam hours will be available and radiation damage studies would make an excellent candidate to conduct in parallel with the nuclear physics program.

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