2014

Exploring a Novel Approach to Technical Nuclear Forensics Utilizing Atomic Force Microscopy

Richard Scot Peeke
College of William & Mary - Arts & Sciences

Exploring A Novel Approach to Technical Nuclear Forensics Utilizing Atomic Force Microscopy

Richard Scot Peeke

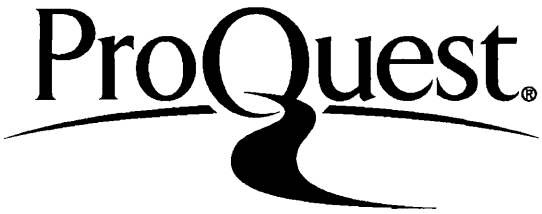
Yorktown, VA

Bachelor of Science, United States Air Force Academy, 1991

A Thesis presented to the Graduate Faculty
of the College of William and Mary in Candidacy for the Degree of
Master of Science

Department of Applied Science

The College of William and Mary
May 2014



ProQuest LLC.
789 East Eisenhower Parkway
P.O. Box 1346
Ann Arbor, MI 48106 - 1346

APPROVAL PAGE

This Thesis is submitted in partial fulfillment of
the requirements for the degree of

Master of Science

Richard Scot Peeke

Approved by the Committee, April 2014

Committee Chair
Assistant Professor Hannes C. Schniepp, Applied Science
The College of William and Mary

Professor Michael J. Kelley, Applied Science
The College of William and Mary

Professor Mark K. Hinders, Applied Science
The College of William and Mary

COMPLIANCE PAGE

Research approved by

Institutional Radiation Safety Committee

Protocol number(s): 13-21

Date(s) of approval: 10 July 2013

ABSTRACT

The collapse of the Soviet Union, proliferation of nuclear materials, and threat of international terrorist organizations propelled the emerging field of Technical Nuclear Forensics (TNF) to the forefront as an international security priority. Technical Nuclear Forensics leverages a multi-disciplinary approach involving law enforcement, intelligence, scientific assets, and the military to perform timely and accurate source attribution through the analysis of nuclear and radioactive materials recovered from illicit storage, interdicted prior to an attack, or collected shortly thereafter. Of utmost importance, is the ability to identify and employ specific markers or signatures that differentiate these materials and reveal details implicating the perpetrators. The purpose of this research is to explore a novel approach to Technical Nuclear Forensics and demonstrate the use of Atomic Force Microscopy (AFM) for pre- and post-detonation analysis of these materials by conducting a comparative *ex-situ* nanoscale morphological and topographical characterization of three specifically selected samples. Trinitite, otherwise known as "Atomsite" or "Alamogordo Glass," was formed during the first detonation of a nuclear weapon, at the Trinity Test Site, Alamogordo, NM, on 16 July 1945. Kharitonchik, an analog of Trinitite, was formed during the Soviet Union's first atmospheric nuclear tests at the Semipalatinsk Test Site, Kazakhstan, on 29 August 1949. In both cases, the crater material, weapon, and test site infrastructure were instantly vaporized in the intense heat of the explosion. The vaporized material was sucked into the rising fireball, cooled, condensed, and then fell to the ground as a melt-glass material. However, Fulgurite, also known as "Desert Glass" or "Petrified Lightning," is naturally formed by lightning strikes of the earth's surface (in this case, the sand dunes at Jockey's Ridge State Park, NC). The lightning instantly melts the sand, silica, or soil and fuses the grains together, forming an amorphous mineraloid. Employing AFM, it is possible to determine the micro-structural properties, geometries, surface roughness, and chemical surface homogeneity of these samples. This initial study intends to lay the groundwork for future research demonstrating the speed, accuracy, and precision of AFM in the determination of provenance.

TABLE OF CONTENTS

ACKNOWLEDGEMENTS

This author wishes to express his appreciation to his faculty advisor and mentor, Professor Hannes C. Schniepp, under whose guidance this research was conducted, for his patience, encouragement, and insight throughout the investigation.

This author is also indebted to the other Master's Thesis Committee members, Professors Mark K. Hinders and Michael J. Kelley, for their careful reading and criticism of the manuscript.

In addition, this author owes a special debt of gratitude to his very supportive lab group partners, especially Mrs. Laura Dickinson and Mr. Sean Koebly, for generously sharing their time, experience, and knowledge, as well as, two very special staff members, Mrs. Lydia Whitaker and Mrs. Rosario Fox.

This author wants to thank the U.S. Army for providing him with an amazing opportunity to pursue his graduate studies at a world-class institution like The College of William and Mary.

Furthermore, this author would like to take a moment to recognize two colleagues from the NASA Langley Research Center Advanced Materials and Processing Brand, Mr. Michael Funk and Mr. Harold Claytor, for their professional expertise and assistance.

Lastly, this author could not have conducted this research without the help of Dr. Byron Ristvet, who sent the Trinitite and Kharitonchik samples from the Defense Nuclear Weapons School, Kirtland AFB, NM, and Superintendant Debo Cox, who sent the Fulgurite samples from Jockey's Ridge State Park, Nags Head, NC...thank you.

I dedicate my research and this thesis to my family...my wonderful wife, my four amazing children, as well as, my steadfast mother and father...for their endless love, support, and encouragement...they are my center, from which, I continually draw hope, inspiration, and the determination to carry on!

LIST OF TABLES

LIST OF FIGURES

1.0 INTRODUCTION

It is a commonly held opinion, within the national and international security communities, that it is not a matter of *if* a terrorist organization decides to use a nuclear or radiological weapon but *when*. This is an exceptionally grave threat because such an attack could cause catastrophic loss of life, extreme economic consequences, profound and lasting social effects, as well as, public loss of confidence in government (Ulicny, 2009). President Barack Obama acknowledged the increased risk and its significance during a 07 April 2009 speech on nuclear weapons in Prague, Czech Republic, when he stated, "in a strange turn of history, the threat of global nuclear war has gone down, but the risk of a nuclear attack has gone up" (Ulicny, 2009). As a counter-response to this increased risk, the September 2006 National Strategy for Combating Terrorism states:

> The rapid identification of the source and perpetrator of an intended or actual WMD attack will enable our response efforts and may be critical in disrupting follow-on attacks. We will develop the capability to assign responsibility for the intended or actual use of WMD via accurate attribution – the rapid fusion of technical forensic data with intelligence and law enforcement information (Ulicny, 2009).

In other words, we will find out who conducted the attack or attempted to conduct an attack using WMD (Weapons of Mass Destruction including Chemical, Biological, Radiological, Nuclear, and High-Yield-Explosive) and we will hold

them accountable by quickly leveraging a combination of scientific analysis, intelligence, and law enforcement in the determination of provenance. The specific term given to this attribution process regarding those cases involving the use of nuclear or radiological materials is Technical Nuclear Forensics. It is regarded by the United States government and the international community as one of the most critical security missions in existence. Therefore, our purpose for this research was to support the Technical Nuclear Forensics mission by exploring the use of a proven, cutting-edge, analytical technique in a novel way. That being said, the goals and objectives for this research were fairly simple and straightforward. First, we wanted to perform some ground-breaking work regarding the potential use of Atomic Force Microscopy (AFM) in Technical Nuclear Forensics. Second, we wanted to demonstrate a unique analytical capability using three specifically selected melt-glass samples representing post-detonation material (these samples include Trinitite, from the first detonation of a nuclear weapon at the Trinity Test Site, Alamogordo, NM; Kharitonchik, from the Soviet Union's first nuclear test at the Semipalatinsk Test Site, Kazakhstan; and Fulgurite (aka "Desert Glass" or "Petrified Lightening"), from Jockey's Ridge State Park, NC). And third, we wanted to develop a way-ahead for future research in the field.

The importance of this type of research cannot be overstated, as the threat is very real. In fact, according to the 2006 International Atomic Energy Agency Nuclear Forensics Support Reference Manual, "illicit trafficking of nuclear

and other radioactive material has been an issue of concern since the first seizures in the early 1990s. By the end of 2004, Member States had confirmed 540 cases, while about another 500 remain unconfirmed" (IAEA, 2006). In a 2006, Scientific American article, entitled, "Thwarting Nuclear Terrorism," Glasser and von Hippel state that, if a well-funded, subnational terrorist organization could obtain as little as 60 kilograms of highly enriched uranium (HEU), then they could construct a crude gun-type device like the World War II "Little Boy" weapon that leveled Hiroshima with a 15 kiloton explosion. The truly frightening revelation from this article is that "more than 50 metric tons (50,000 kilograms) of highly enriched uranium" are located at nearly 140 "often poorly secured" civilian nuclear research facilities worldwide. Furthermore, in a November 2013 National Journal Magazine article entitled, "A Glimpse at the Next Bin Laden," Michael Hirsh wrote,

> With the exception of Egypt—where the military has cracked down on the Muslim Brotherhood—the Arab Spring uprisings have opened up huge swaths of ungoverned territory in Muslim nations that once cooperated with Washington against terrorism. The toppling of strong autocratic leaders has led not to secular democracy but to fractionalization, allowing some Islamist groups to seize territory in which they might host terrorists cells in the way the Taliban welcomed bin Laden. "There are at least 25 failed states in the world, an unprecedented number," says Pascal Boniface, head of the Paris-based Institute for International and Strategic Relations. They stretch from Yemen and Somalia to Syria and Libya and Iraq (Hirsh, 2013).

In addition, Forecasting International Inc. published an article, on behalf of The National Intelligence University, Office of the Director of National Intelligence, and the Center for Strategic Leadership - U.S. Army War College, written by Dr. Marvin J. Cetron and Mr. Owen Davies, in February 2008, entitled, "55 Trends Now Shaping the Future of Terrorism." It stated the following:

> We take it for granted that the elite among tomorrow's terrorists will have more than plastic explosives with which to make their point. They will have nuclear weapons...Wherever secular government is weak, it might easily be replaced by a much stronger and more virulently anti-American theocracy with leaders drawn straight from the terrorist movement...From here on out, nuclear terrorism is a realistic threat...This is clearly the single most important trend for terrorism (Cetron & Davies, 2008).

Lastly, during a 26 July 2011 United States Government Accountability Office testimony before the U.S. House of Representatives, Committee on Homeland Security, Subcommittee on Cybersecurity, Infrastructure Protection, and Security Technologies, David C. Maurer (Director, Homeland Security and Justice) and Gene Aloise (Director, Natural Resources and Environment) were quoted, as follows:

> Terrorists smuggling nuclear or radiological material into the United States could use these materials to make an improvised nuclear device or a radiological dispersal device (also called a "dirty bomb"). The detonation of a nuclear device in an urban setting could cause hundreds of thousands of deaths and devastate buildings and physical infrastructure for miles. While not as

> damaging, a radiological dispersal device could nonetheless cause hundreds of millions of dollars in socioeconomic costs as a large part of a city would have to be evacuated.—and possibly remain inaccessible.—until an extensive radiological decontamination effort was completed (Maurer & Aloise, 2011).

So the bottom line is that this is a very serious national and international problem requiring immediate attention and the consideration of a variety of analytical techniques to include novel approaches. Our first step in this project was to determine the extent of the current research pertaining to Technical Nuclear Forensics and AFM. Our literature review produced dozens of articles addressing the threat of nuclear terrorism and the employment of Technical Nuclear Forensics as a counter to this threat. This thesis lists no less than seventeen papers pertaining to nuclear forensics in general, nine papers focused on the specific analysis of Trinitite, four papers addressing the investigation of the soil samples in and around the Semipalatinsk Nuclear Test Site, and one paper commenting on the formation and morphology of Fulgurite.

A summary of the relevant information regarding nuclear forensics in general reveals many consistencies throughout the literature. Nearly all of the papers acknowledge the importance of determining specific forensic signatures, markers, or parameters (Joint Working Group, 2008; Kristo & Tumey, 2013; Mayer, Wallenius & Fanghanel, 2007; Stanley, Stalcup & Spitz, 2013). Similarly, all seem to agree that there is a significant need for a comprehensive international nuclear forensic database including technical reference material

pertaining to, for example, nuclear fuels, uranium ores, weapon materials, commercial reactors, and research reactors (Mayer et al., 2007). According to Stanley et al. (2013), some of the significant challenges in the field of Technical Nuclear Forensics include international cooperation and collaboration, the current and future availability of qualified expertise, as well as, research and development in key technical areas like automated, field-deployable, analytical instrumentation. Added to this, Mayer et al. (2007) mention safely working with radioactive materials, chain-of-custody considerations, and the proper handling of evidence as additional challenges confronting the Technical Nuclear Forensic community.

In terms of the various analytical techniques employed, the literature reveals a wide variety of approaches, including radiochemical separation, optical microscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray fluorescence (XRF), neutron activation analysis (NAA), and a full array of counting techniques, to include alpha spectrometry, gamma spectrometry, accelerator mass spectrometry (AMS), resonance ionization mass spectrometry (RIMS), secondary ionization mass spectrometry (SIMS), thermal ionization mass spectrometry (TIMS), electron-dispersive X-ray spectroscopy (EDX), and multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS), to determine such things as isotopic ratios, chemical impurities, age of material, microstructures, and macroscopic parameters (Joint Working Group, 2008; Kristo et al., 2013; Mayer et al., 2007; Stanley et al., 2013; Tuniz &

Hotchkis, 2004). However, none of these references specifically noted the use of AFM in Technical Nuclear Forensic research.

A summary of the relevant information regarding the nine studies involving the specific analysis of Trinitite also reveals many consistencies throughout the literature. Salter and Harley (1965) employed gamma spectrometry to determine the presence of ^{60}Co, ^{137}Cs, and ^{152}Eu. Similarly Mercer, Hypes, and Saey (2010) used gamma spectrometry to determine that "the radioisotopes ^{152}Eu ($T_{1/2}$=12.7 years) and ^{154}Eu ($T_{1/2}$=8.5 years) were generated by neutron irradiation of natural europium in the soil, ^{137}Cs ($T_{1/2}$=30.1 years) was a fission product from the nuclear detonation, and ^{60}Co ($T_{1/2}$=5.3 years) was produced mainly by irradiation of steel structures." In 2006, a well-known study used a combination of radiochemistry, alpha spectrometry, gamma spectrometry, and beta counting...to determine specific activities of the "fission products ^{90}Sr and ^{137}Cs, activation products ^{60}Co, ^{133}Ba, ^{152}Eu, ^{154}Eu, ^{238}Pu, and ^{241}Pu, the remains of the nuclear fuel ^{239}Pu and ^{240}Pu, as well as natural radionuclides ^{40}K, ^{232}Th, and ^{238}U and their progeny" (Parekh, Semkow, Torres, Haines, Cooper, Rosenburg, & Kitto). The results of this study suggested that various radionuclides were "volumetrically incorporated" into the molten Trinitite glass, including "fission products from ^{239}Pu fission; activation products from neutron reactions with the nuclear fuel, the materials used in construction of the Gadget, and the local GZ soil; the remnants of unburned nuclear fuel; as well as radionuclides of natural origin in the soil." In a similar fashion, Fahey, Ritchie,

Newbury, and Small (2010) utilized a suite of analytical tools including alpha spectroscopy, gamma spectroscopy, electron microprobe (e-probe), scanning electron microscopy (SEM), light microscopy, X-ray florescence (XRF), autoradiography, and secondary ion mass spectrometry (SIMS) to conduct one of the first studies considering the forensic analysis of post-detonation nuclear debris in an attempt to retroactively understand the device that produced the explosion. They concluded that "bulk analytical methods are unable to adequately tease apart populations of end-member components derived from distinct starting materials, whereas microscopic methods can detect lower levels of materials due to localized microconcentrations and spatial relationships." Furthermore, with regards to Trinitite, they stated that "the visible heterogeneity of the sample near the surface indicates that the material was not completely mixed, forming a homogeneous glass, but rather specific signatures have been preserved in the heterogeneity of the glassed material." This observation bodes well for those seeking to attribute an attack to a specific organization by examining the bomb material and debris. Using autoradiography on thin sections and scanning electron microscopy energy dispersive X-ray spectroscopy (SEM-EDX) analysis and alpha spectrometry on fine powder samples, Belloni, Himbert, Marzocchi, and Romanello (2011) proposed a "detailed model of the overall mechanism leading to Trinitite formation" to include suggesting a two-stage process. Finally, the Bellucci research team (from the University of Notre Dame) has published multiple papers regarding their analysis of Trinitite. The first of these studies that we reviewed involved the use of scanning electron microscopy

(SEM), backscatter electron imaging (BSE), and energy dispersive X-ray spectroscopy (EDS) to document "the 3D morphology of Trinitite-hosted metallic inclusions and the first observations of alloys consisting primarily of Pb, Ta, Ga, and W" presumably from the weapon components (Bellucci & Simonetti, 2012). Bellucci et al. assert that understanding the local geology is essential to differentiating bomb components from the local materials. Furthermore, based on chemical compositions, it is the metals (Fe, Pb) and radionuclides (Pu, U, and related activation/fission products) that provide the greatest contrast. Their results emphasize the need for "non-destructive and micron- (sub-micron-) scale characterization" prior to bulk dissolution methods, reinforcing the conclusions espoused by Fahey et al (2010). The second study involved the largest sample size to date and was conducted using gamma spectroscopy to determine the distribution and behavior of various radionuclides associated with the Trinity nuclear test; it suggested that "the energy produced in the Trinity test was derived from the fission of both ^{239}Pu and ^{235}U" (Bellucci, Wallace, Koeman, Simonetti, Burns, Kieser, & Walczak, 2013). The third and fourth studies used laser ablation multi-collector inductively coupled plasma mass spectrometry (LA-MC-ICP-MS) on polished thin sections to determine U isotopic concentrations and Pb isotopic concentrations, respectively (Bellucci, Simonetti, Wallace, Koeman, & Burns, 2013). Based on this research, Bellucci et al. concluded that the "best forensic evidence to determine the provenance of a nuclear device lies in the isotopic composition of heavy metals (Pb, Pu, and U)."

Our literature review suggests that AFM could be useful with regards to imaging and mapping the various aforementioned inclusions. However, before discussing the specifics pertaining to our research methodology, results, discussion, and conclusion, the next two sections will briefly address some background information regarding Technical Nuclear Forensics, as well as, some background and scientific theory pertaining to AFM.

1.1 Technical Nuclear Forensics (TNF) Background

According to the National Technical Nuclear Forensics Center, which reports to and through the Domestic Nuclear Detection Office, under the Department of Homeland Security, Technical Nuclear Forensics is defined as:

> The collection, analysis, and evaluation of pre-detonation (intact) and post-detonation (exploded) radiological or nuclear materials, devices and debris, as well as, the immediate effects created by a nuclear detonation...Technical Nuclear Forensics, combined with law enforcement and intelligence information, support nuclear attribution – the identification of those involved with planned or actual attacks using radiological or nuclear weapons or materials (NTNFC, n.d.).

Therefore, Technical Nuclear Forensics serves as a deterrent against proliferation, nuclear terrorism, illicit trafficking, and illegal dumping. This point is further reinforced by the Joint Working Group of the American Physical Society and the American Association for the Advancement of Science when they state in their paper entitled, "Nuclear Forensics: Role, State of the Art, Program

Needs" that a "believable attribution capability may help to discourage behavior that could lead to a nuclear event" (2008). Technical Nuclear Forensics employs a "whole of government" approach involving inter-agency assets from the Departments of Homeland Security, Energy, Defense, Justice, State, and the Office of the Director of National Security. Technical Nuclear Forensics seeks to collect, analyze, and evaluate specific samples in order to identify physical, chemical, elemental, and isotopic nuclear forensic signatures based on the production, processing, or source material, helping investigators to determine the age, origin, or intended use of the material (see Fig. 1).

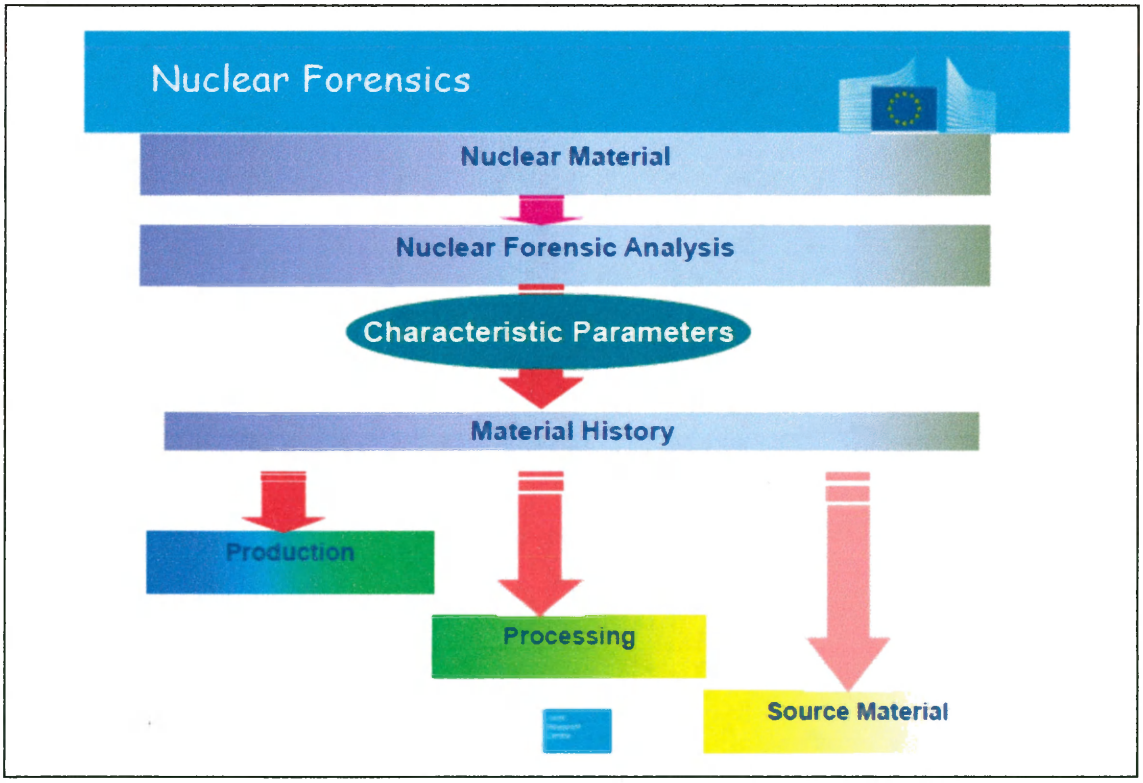


Figure 1: From Nuclear Forensics – A Methodology to Reveal the Past of Nuclear Materials by Maria Wallenius, Klaus Mayer, and Zsolt Varga of the European Commission, Joint Research Center, Institute for Transuranium Elements, Karlsruhe, Germany (Wallenius, n.d.).

On the operational side, Figure 2 (below) provides a simplified illustration of a post-detonation, inter-agency, ground-sampling mission, concept of employment graphic. National Technical Nuclear Forensic (NTNF) mission assets support the United States Government (USG) attribution process by providing rapid collection, analysis, and assessment so that National Command Authorities can make informed decisions in a timely fashion. A Forward Operating Base (FOB) is quickly established, from which sample teams can run collection missions. Team 1 attempts to collect volatile samples, including dust and fine or light particles that float for greater distances. Team 2 attempts to collect refractory samples, including dense daughter products, unused fuel oxides, and other small or spherical particles that condense out early. The Aerial Radiation Detection Identification and Mapping System (ARDIMS) supports these collection missions by providing real-time aerial surveys of radiological ground deposition. Personnel, equipment, and samples are processed through the hotline for decontamination. Initial presumptive analysis is conducted in the DOE Triage tent, which contains a number of field-deployable, analytical instruments used for the rapid identification and characterization of the samples (at the present, atomic force microscopy is not included in this suite of analytical capabilities). Samples are then transferred to national laboratories for more extensive confirmatory analysis, while a strict chain of custody is maintained to ensure proper evidence protection and preservation. Experts agree that a program to develop, test, and manufacture advanced, specialized, automated, field-deployable equipment, as well as, sophisticated laboratory instruments for

the purpose of conducting rapid and accurate Technical Nuclear Forensic measurements is sorely needed in the United States (Joint Working Group, 2008).

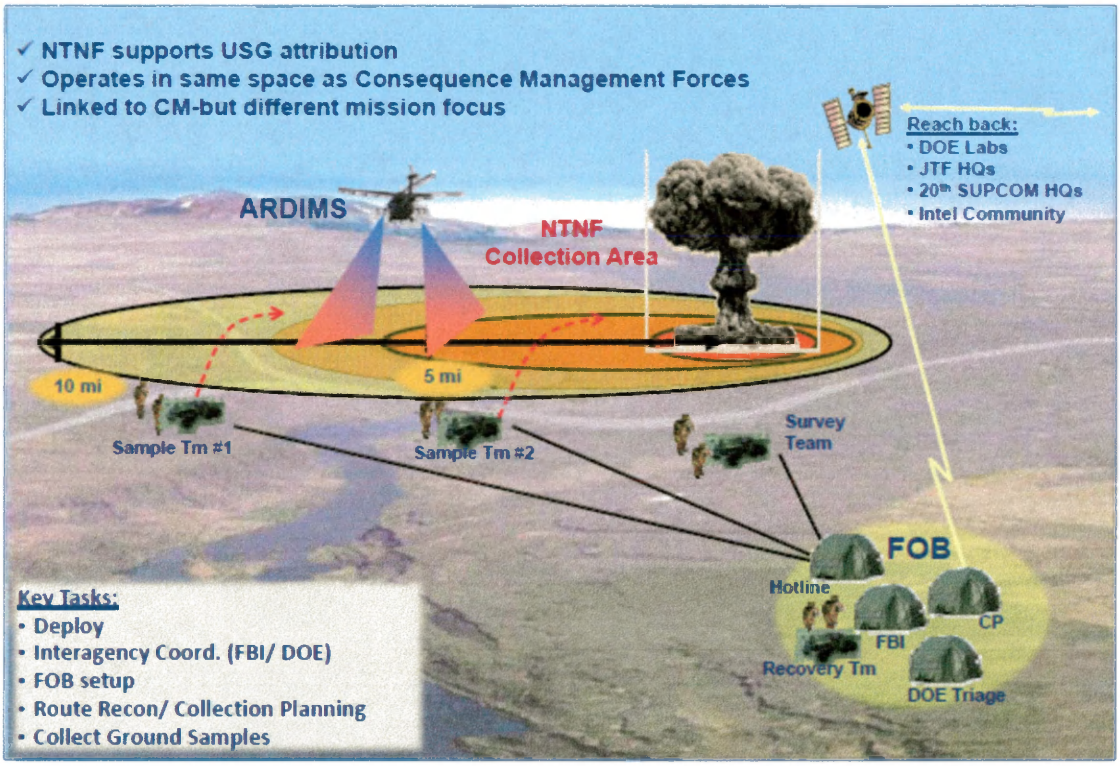


Figure 2: From National Technical Nuclear Forensics Mission Brief Slides (Esce, 2013).

1.2 Atomic Force Microscopy (AFM) Background

"Atomic force microscopy is an amazing technique that allows us to see and measure surface structure with unprecedented resolution and accuracy" all the way down to the arrangement of individual atoms (Eaton & West, 2010). Unlike an optical or electron microscope, AFM physically feels the sample

surface with a sharp probe (or tip), building a map of the surface height (see Fig. 3).

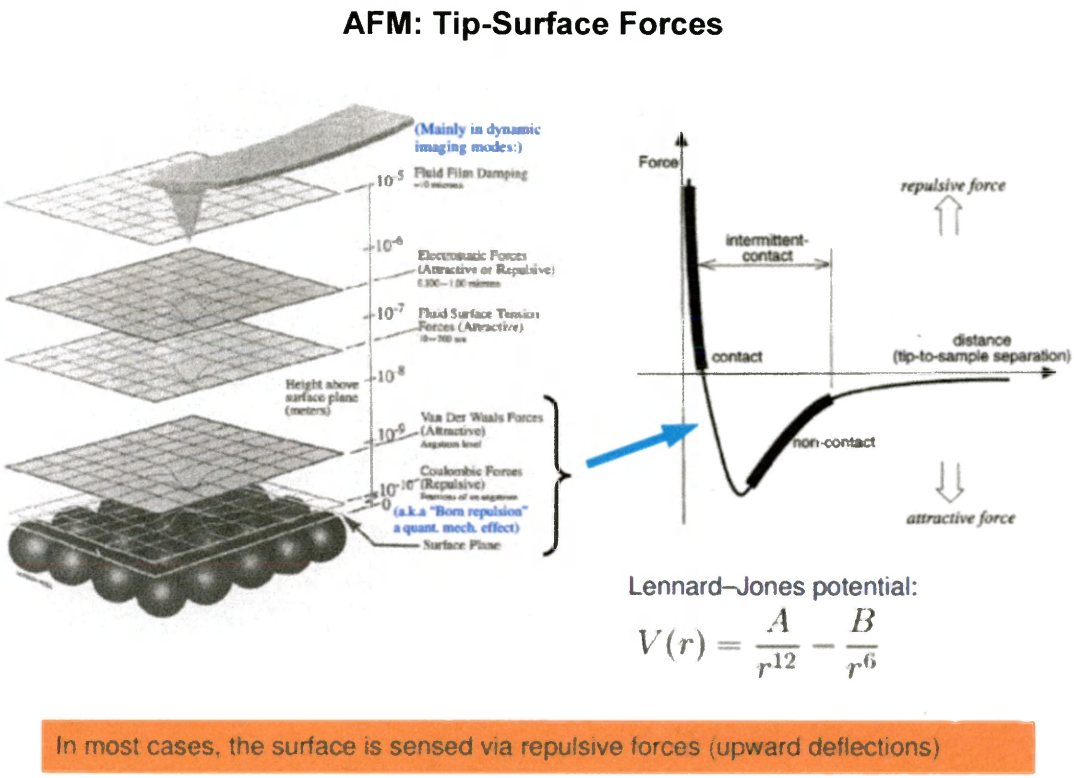


Figure 3: AFM relies on tip-surface interactions characterized by the Lennard-Jones Potential between attractive Van der Waal forces and repulsive forces due to the Born Quantum Mechanical Effect (Veeco, n.d.).

AFM provides exceptionally high resolution in the Z-direction, well below the 250 nm resolution limit of optical microscopes. In fact, AFM sensitivity can measure down to 0.01 nm (or 0.1 angstrom)...roughly 1/10th the diameter of an individual atom. Thormann, Pettersson, and Claesson (2009) state that probe-surface interactions "in the range from a few piconewtons to several micronewtons can be detected." AFM produces "real" topography versus pseudo-topography from back-scattering. AFM allows the use of different modes

and probes to map different qualities, characteristics, and properties like material modulus and stiffness. AFM makes it possible to image inclusions or impurities, that are not otherwise visible, via lateral friction force scans and other modalities. AFM requires relatively simple, quick, non-destructive sample preparation under ambient conditions and produces equally fast results. Lastly, the instruments themselves occupy a relatively small footprint and require little in the way of support infrastructure other than a clean, quiet, and vibration-free work space with electrical power.

The three basic AFM modes include contact, semi-contact, and non-contact. Semi-contact and non-contact are often referred to as dynamic mode AFM. This initial research was conducted using three variations of contact mode AFM (CM-AFM) as follows in Figures 4 through 6.

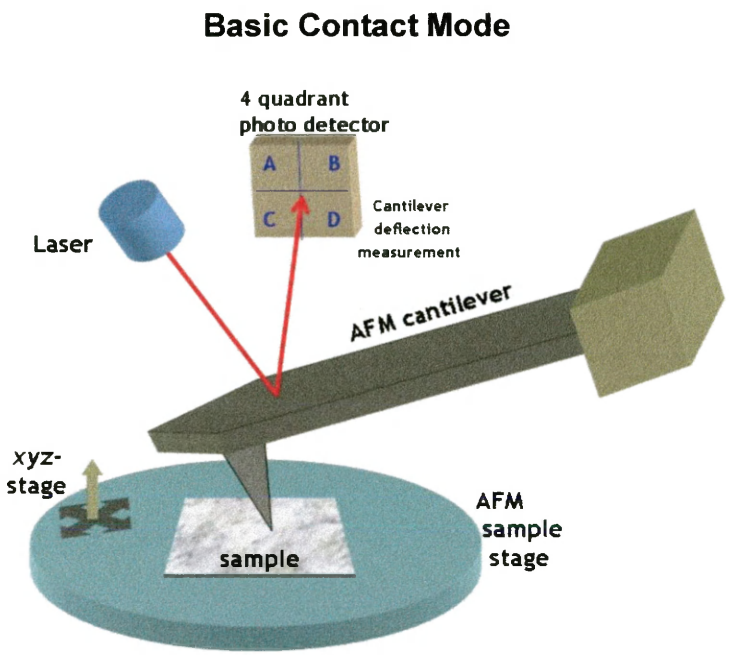


Figure 4: Basic Contact Mode AFM utilizes a sharp probe (or tip) at the end of a flexible cantilever to interrogate the sample surface in a raster graphic pattern. The tip-surface interaction is measured and recorded via the deflection of a laser off of the cantilever onto a photodiode as height data (Yashvant, 2013)

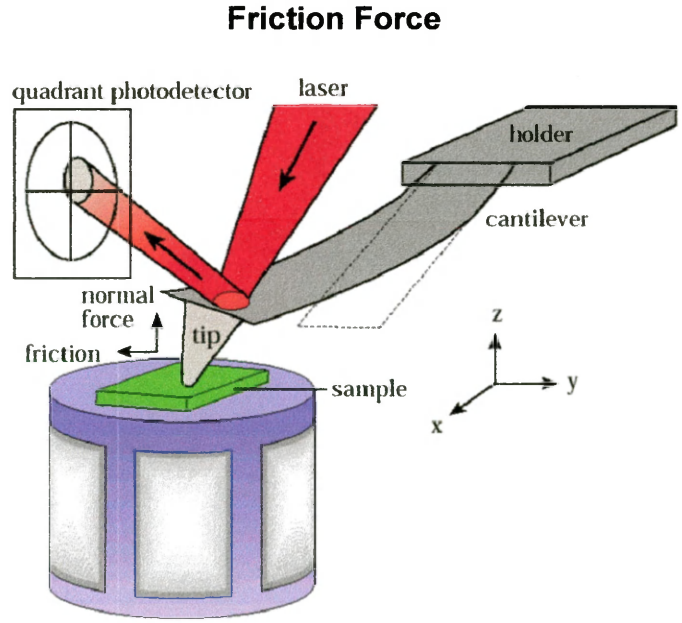


Figure 5: Friction Force scans are conducted using the same contact mode set up as before, however, in this case, the system measures and records the torsion of the flexible cantilever as the tip moves laterally across the sample surface, indicating different material friction force coefficients and chemical compositions (Friction Force Graphic, 2013).

Force Spectroscopy

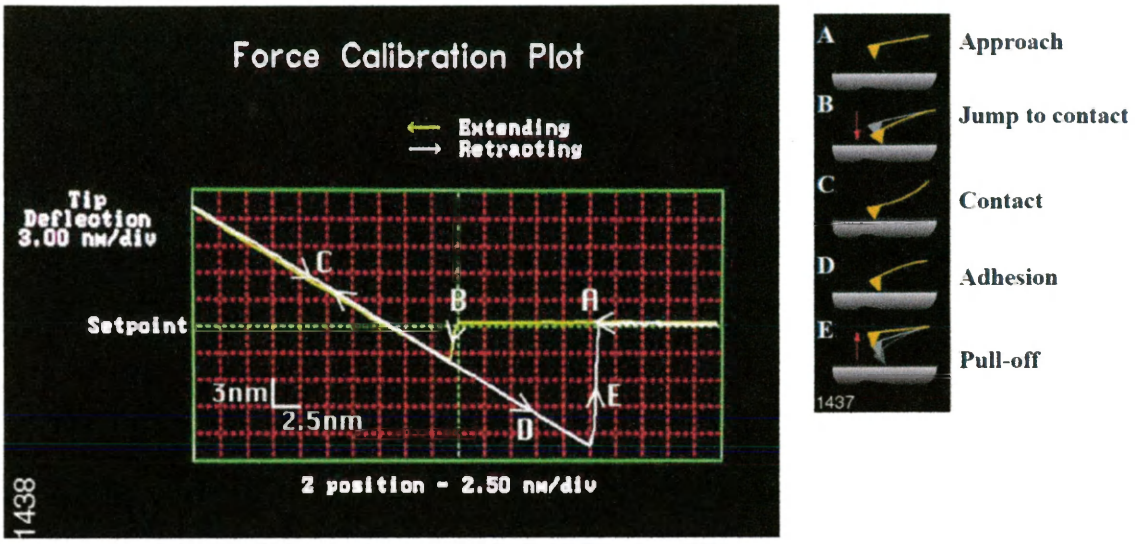


Figure 6: Force Spectroscopy is also conducted using the contact mode set up. For these measurements, the tip is approached vertically towards the sample, while the cantilever deflection is monitored. Then the tip is retracted towards the initial position. The cantilever deflection is plotted versus the vertical piezo displacement for each direction (Veeco, 2000).

Each of the three contact mode AFM variations used a special silicon nitride (SiNi) probe with a micro-fabricated cantilever of monolithic design (see Fig. 7).

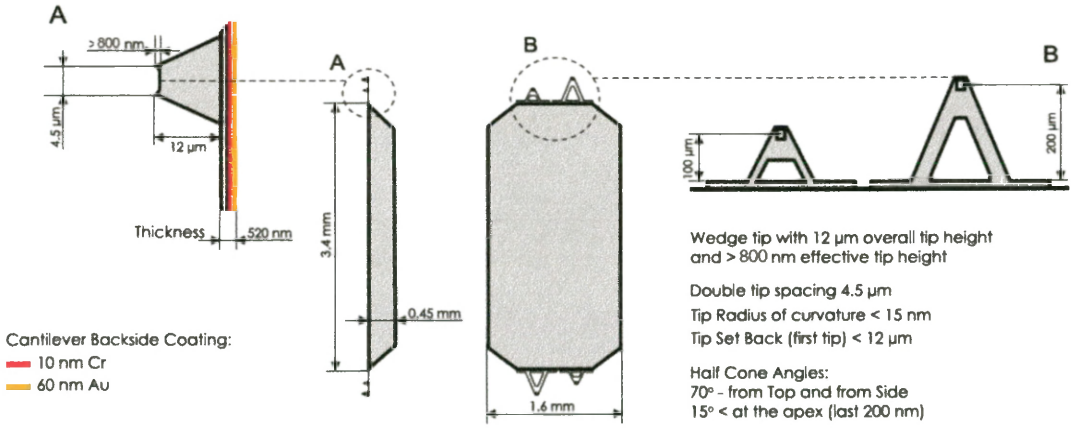


Figure 7: Budget Sensors SiNi Soft Contact Mode AFM Probe with 4 Silicon Nitride Cantilevers (triangular, 2 different lengths); Cantilever lengths: 100 µm and 200 µm; Coating: 70 nm thick Gold/Chromium (SiNi probe, n.d.).

2.0 RESEARCH METHODOLOGY

The Institutional Radiation Safety Committee at The College of William and Mary approved all procedures associated with this research via Protocol Approval Number 13-21. As stated in the approved protocol, the purpose of this research was to conduct a comparative analysis and characterization of Trinitite, Kharitonchik, and Fulgurite melt-glass samples utilizing AFM in order to demonstrate this technology's application to the field of technical nuclear forensics. Since two of the three samples were produced during nuclear tests, it was determined that all reasonable and appropriate safety measures and requirements would be followed.

Therefore, we reviewed The College of William and Mary's Radiation Safety Manual to ensure that this research was in compliance with all specified requirements. Furthermore, we successfully completed The College of William and Mary's three hour Radiation Safety Course. In addition, the samples were brought to The College of William and Mary's Radiation Safety Officer for initial monitoring. At 13-22 μR/hr (at near contact) for the Trinitite and 25-30 μR/hr (at near contact) for the Kharitonchik, the samples were determined to be at or below twice background for the Integrated Science Center and well below the minimum survey meter sensitivity of 0.1 mREM/hr specified in The College of William and Mary's Radiation Safety Manual. Lastly, despite the fact that these experimental samples fell below the threshold for monitoring under The College

of William and Mary's Radiation Safety Program, we still followed the ALARA (As Low As Reasonably Achievable) Principle, essentially using procedures and engineering controls based upon practical, sound, radiation protection practices to minimize occupational doses and public exposure. All samples were appropriately labeled and stored in McGlothlin Street Hall Lab #307 behind a cipher locked door. The samples were imaged in McGlothlin Street Hall Lab #39, which was also locked for security purposes. Full disclosure was made, on a need to know basis, to include the opportunity to submit a Declaration of Pregnancy. The radiation safety concepts of time, distance, and shielding were employed at all times. Lastly, reasonable lab practices were enforced to include the use of protective gloves, protective eyewear, frequent hand-washing, frequent cleaning of the lab area, and no eating, drinking, smoking, or application of cosmetics in the lab area.

The following subsections will address details regarding sample selection and acquisition, the project planning matrix, and the lab set-up, to include the specific equipment used during the investigation.

2.1 Sample Selection and Acquisition

Trinitite, otherwise known as "Atomsite" or "Alamogordo Glass," was formed during the first detonation of a nuclear weapon, at the Trinity Test Site, Alamogordo, NM, on 16 July 1945 (Trinitite, 2013). The test device, nicknamed “The Gadget,” was a 20 kt implosion-design plutonium fission device. Trinity was an atmospheric test conducted atop a 100 ft steel tower. The detonation created

a crater in the Arkosic sand (composed of quartz grains and feldspar) that was 3.0 m deep and 340 m wide, ushering in the beginning of the "Atomic Age." The blast temperature was at least 1470 °C and produced a mushroom cloud approximately 12.1 km high (Trinity Nuclear Test, 2013). The crater material, the weapon, and the test site infrastructure were instantly vaporized in the intense heat of the explosion. The vaporized material was sucked into the rising fire ball, cooled, condensed, and fell to the ground as a light green melt-glass material (see Fig. 8).

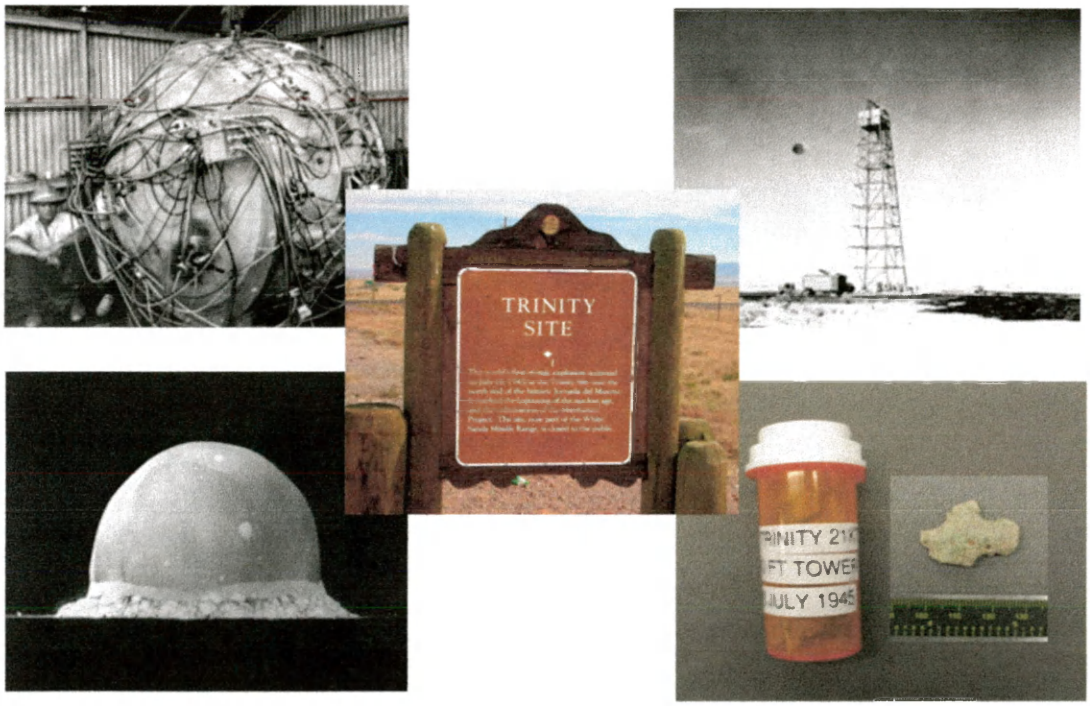


Figure 8: The Trinitite melt-glass samples (appox. 33 g) collected from the Trinity Nuclear Test Site, Alamogordo, NM. They were sent as a gift by Dr. Byron L. Ristvet, Assistant for Nuclear Matters (DTRA/RD-CXT), on 03 June 2013, via conventional nonhazardous shipping (Trinity Test Site, n.d.).

Kharitonchik, an analog of Trinitite, was formed during the Soviet Union's first atmospheric nuclear tests at the Semipalatinsk Test Site, Kazakhstan, on 29 August 1949. "RSD-1" (aka JOE-1 by U.S. intelligence) was a 22 kt implosion-design plutonium fission device (very similar to the "Gadget" and the "Fat Man" weapon dropped over Nagasaki, Japan). "First Lightening," as it was code-named, was also an atmospheric test conducted from atop a tower. The subsequent detection of the resulting mushroom cloud and public disclosure of the test accelerated the beginning of the "Cold War" (RSD-1, 2013). This porous black melt-glass material was formed in much the same way as Trinitite and was named after Dr. Yulii Borisovich Khariton (seated next to the weapon in Fig. 9), a leading Russian nuclear weapons scientist (Trinitite, 2013).

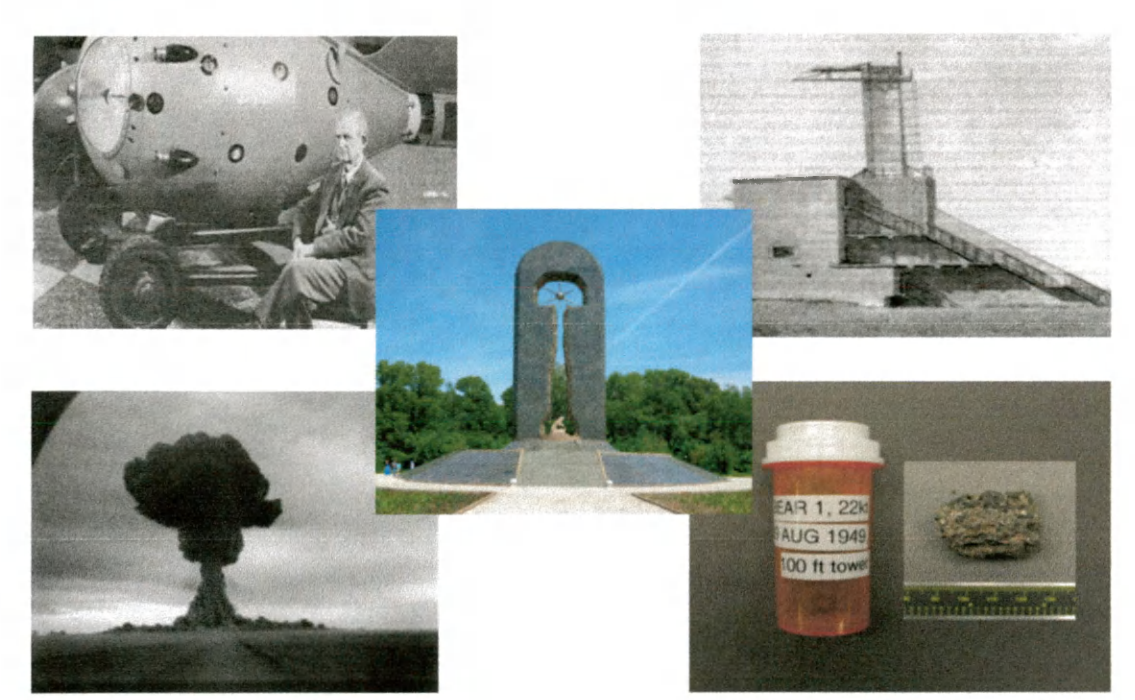


Figure 9: The Kharitonchik melt-glass samples (approx 30 g) were collected from the Semipalantinsk Nuclear Test Site, Kazakhstan. They were sent as a gift by Dr. Byron L. Ristvet, Assistant for Nuclear Matters (DTRA/RD-CXT), on 03 June 2013, via conventional nonhazardous shipping (Semipalantinsk Nuclear Test Site, n.d.).

Fulgurite (from the Latin *fulgar* meaning thunderbolt), also known as "Desert Glass" or "Petrified Lightning," is naturally formed in sand, silica, or soil by lightning strikes of the earth's surface (in this case, the sand dunes at Jockey's Ridge State Park, NC). The lightning, at a minimum temperature of 1,800 °C, instantly melts the sand, silica, or soil and fuses the grains together, forming an amorphous mineraloid in the form of the hollow glass tubes pictured in Figure 10 (Fulgurite, 2013).

Figure 10: The Fulgurite melt-glass samples (approx 4 g) were collected at Jockey's Ridge State Park, Dare County, NC. They were sent as a gift by Park Ranger Debo Cox, Superintendent, Jockey Ridge State Park, on 01 May 2013, via conventional nonhazardous shipping (Jockey's Ridge State Park, n.d.).

The Trinitite and Kharitonchik samples were specifically selected because they represented "post-detonation" melt-glass material from well-known and

documented events. The Fulgurite, on the other hand, was specifically selected because it was formed by a natural process similar to the anthropogenic process caused by a nuclear detonation but without the radiological component. As such, it represented a non-radioactive experimental control material for comparison purposes.

2.2 Project Planning Matrix

When conducting analysis using multiple types of microscopy, on multiple samples, in multiple locations, using multiple sample preparation techniques, and multiple scanning dimensions, it helped to use a planning matrix like the one reflected in Figure 11 to keep organized.

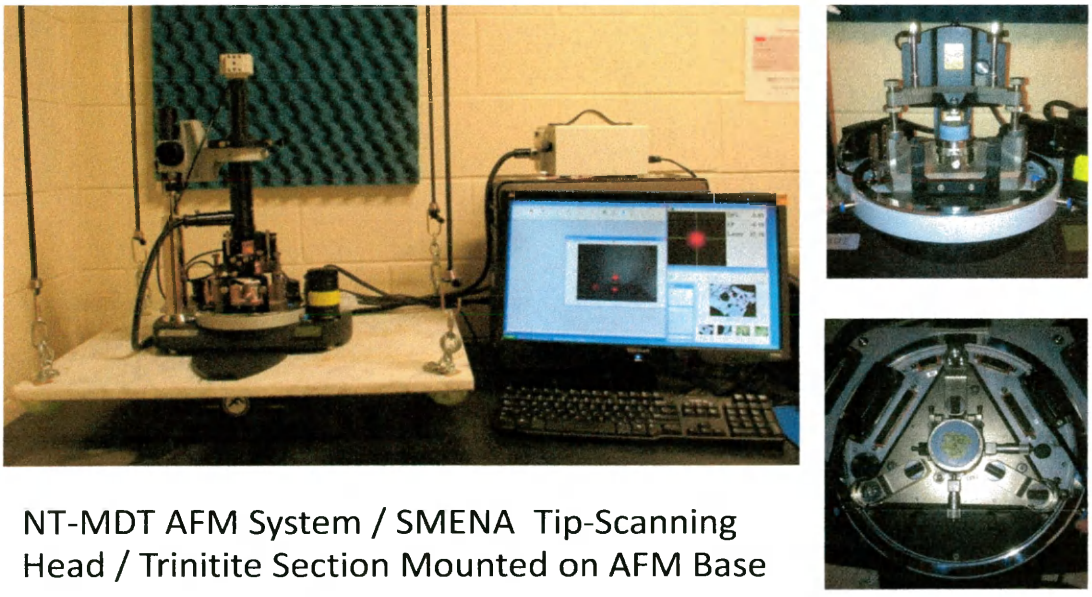
Peeke's Project Planning Matrix (2/26)

SAMPLE TYPE & PREP	ACTIONS														
				Canon	Nikon		Olympus						AFM Contact Height & LFF		
	SP	LZ	Name	1x	1x	3x	5x BF	8x DF	20x BF	32x DF	40x BF	64x BF	1 um	5 um	10 um
Trinitite:															
1. Section - Natural															
2. Section - Polished	x	1	Crossroads	25-Feb	25-Feb	25-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	21-Feb	21-Feb	21-Feb
		2	Seam Below	25-Feb	25-Feb	25-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	21-Feb	21-Feb	21-Feb
3. Epoxy - Natural															
Kharitonchik:															
4. Section - Natural															
5. Section - Polished	x	1	Red Bean	25-Feb	25-Feb	25-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	20-Feb	20-Feb	20-Feb
		2	Blk Pyramid	25-Feb	25-Feb	25-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	21-Feb	21-Feb	21-Feb
6. Epoxy - Natural															
Fulgurite:															
7. Section - Natural															
8. Section - Polished															
9. Section - Polished	x	1	Profile	25-Feb	26-Feb	26-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	24-Feb	24-Feb	24-Feb
		2	Steam Iron	25-Feb	26-Feb	26-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	19-Feb	24-Feb	24-Feb	24-Feb
10. Epoxy - Natural															

Figure 11: Project Planning Matrix.

2.3 Lab Set-Up and Equipment

The AFM Lab at The College of William and Mary consists of two extremely sensitive AFM instruments (the NT-MDT AFM and the Bruker AFM) and is located in the basement of McGlothlin Street Hall (Lab #39) in order to minimize interference due to structural or noise vibration. In fact, the individual instruments rest on a platform suspended from the roof by bungee cords in order to further dampen structural vibrations. Likewise, the walls of the AFM Lab have acoustic foam on them to help dampen interior noise vibrations. We employed the NT-MDT AFM System, equipped with the SMENA Tip-Scanning Head and built-in 100μm x 100μm piezo, in contact mode (CM), utilizing a Budget brand silicon nitride (SiNi) soft AFM probe to produce all of the AFM scans used in this research, as depicted in Figure 12 below.



NT-MDT AFM System / SMENA Tip-Scanning Head / Trinitite Section Mounted on AFM Base

Figure 12: AFM Lab Set-Up and Equipment.

The large image in Figure 12 displays the NT-MDT NOVA software user interface screen and the four quadrants of the photodiode (used for aiming the laser), the optical image of the sample (used to help re-acquire the desired scanning location), as well as, the live camera feed, zoomed in on the cantilever (used to monitor the tip, as it approaches the sample surface). Figure 13 (below) shows close-up images of the soft triangular SiNi contact mode cantilevers "approached" to the designated landing zones for each sample type. The tip-surface interaction is measured and recorded via the deflection of the laser off of the cantilever and onto the photodiode. The images were taken using the organic NT-MDT NOVA software and reflect viewing windows of approximately 3.5 mm X 4.5 mm. We prepared the images for this report using GIMP (GNU Image Manipulation Program), an open-source professional cross-platform raster graphics editor.

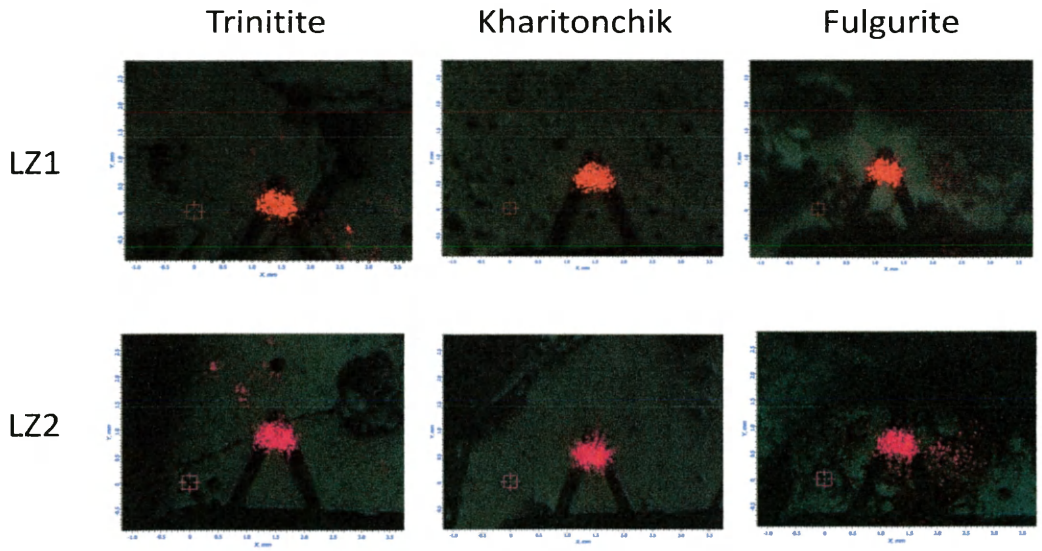


Figure 13: Triangular SiNi contact mode cantilevers "approached" to the designated landing zones for each sample type.

3.0 RESULTS

In accordance with the Project Planning Matrix, sample preparation was the first action to be taken. Through a process of trial and error, various sample preparation techniques were attempted. It was determined that "polished" sample "sections" were preferable to "sections" exposing a sample's "natural" surface or a sample mounted in "epoxy" with the "natural" surface exposed, as addressed further in subsection 3.1. The next action involved optical microscopy of each of the various sample types (sections). This step proved to be extremely valuable in terms of identifying desirable scanning locations and reacquiring those same locations in the AFM lab, as detailed in subsection 3.2. Following optical microscopy, contact mode AFM scans were produced. The height signal and friction signal images were measured and recorded for each of the three sample types (or sections), at each of the two pre-designated LZs, for three different scan range sizes (1 μm x 1 μm, 5 μm x 5 μm, and 10 μm x 10 μm). The 5 μm x 5 μm height signal and friction signal scans are displayed in subsection 3.3, along with Table 1, reflecting the respective contact mode AFM scanning parameters. In addition, three-dimensional (3D) topographical surface images were constructed for each of the three sample types (or sections), at each of the two pre-designated LZs, for each of the 5 μm x 5 μm scans, as displayed in subsection 3.4. Lastly, force curves were taken for each of the three sample types (or sections), at each of the two pre-designated LZs, for each of the 5 μm x

5 μm scans. For each 5 μm x 5 μm scan, a center point force curve and a 5 x 5 grid of force curves were measured and recorded. For qualitative comparison purposes, only the center point force curve for each 5 μm x 5 μm scan is displayed in subsection 3.5. Lastly, Table 2 reflects the relevant input parameters for each of the displayed force curves.

3.1 Sample Preparation

We attempted a few different sample preparation techniques utilizing epoxy cement to mount sample specimens to various substrates like mica or metal pucks for subsequent optical microscopy and scanning with the AFM. However, these attempts produced mixed results, primarily due to excessive sample tilt and surface angulation during the AFM scans. Eventually, we adopted a sample preparation technique utilized by the NASA Langley Research Center (LRC) Advanced Materials and Processing Branch. This technique is similar to the one employed by Belloni et al. (2011) to prepare thin sections for autoradiography during their investigation of radionuclide incorporation and distribution in Trinitite. We "potted" the samples in an acrylic "puck" using the Buehler VariDur Powder High Performance Mounting System together with Buehler Ultramount Hardener producing a blue mineral filled acrylic cast after a 15-30 minute cure time at room temperature. Due to the initial size of the Kharitonchik nugget-like sample, we used the LECO CM-24 Cut-Off Machine to bisect it prior to potting. Then we used the Buehler IsoMet Low Speed Saw to section each of the samples. Lastly, we used the Buehler EcoMet-4 Variable

Speed Grinder-Polisher at 120 rpm to sand and polish the samples, using four different 12-inch, cloth-backed, sanding and polishing discs attached to the wheel via pressure-sensitive adhesive (PSA). The CAMI (Coated Abrasive Manufacturers Institute) ratings for these discs were 400 and 600 grit for sanding, while the polishing procedure used 800 and 1200 grit. The samples were sanded and polished, applying them by hand to the rotating discs, in sequence, starting with the lowest grit to the highest grit, under a constant flow of water. The equivalent average particle diameters in micrometers are 23.6 μm, 16.0 μm, 12.2 μm, and 6.5 μm, respectively (Sandpaper, n.d.). Each of the sections were prepared in the same manner and measured approximately 30-40 mm in diameter. This sample preparation technique solved the problem of sample tilt and surface angulation, while providing a stable, user-friendly platform from which to conduct optical microscopy and perform the three variations of contact mode AFM mentioned previously (see Fig. 14 below).

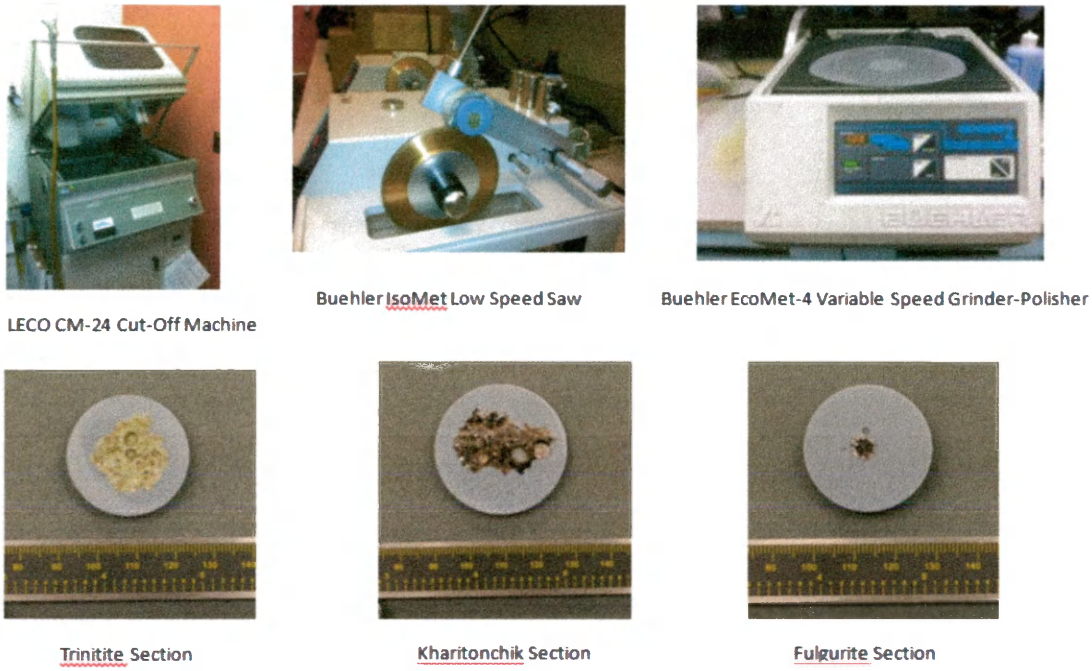


Figure 14: Sample preparation at the NASA Langley Research Center Advanced Materials and Processing Branch.

3.2 Optical Microscopy

We performed optical microscopy on each sample section in the Nanomaterials and Imaging Lab, McGlothlin Street Hall #307, using our Nikon SMZ800 Zoom Stereomicroscope and our Olympus IX-71 Inverted Optical Microscope. Optical microscopy was essential to the selection of desirable locations for AFM scanning and subsequent attempts to reacquire those same pre-designated locations in the AFM lab downstairs. The next three figures (Fig. 15-17) depict optical images of each sample type with two designated landing zones (LZ) per sample type. LZ 1 and LZ 2 represent locations where we intended to conduct an AFM scan based on surface characteristics like flatness,

smoothness, or an interesting topographical feature such as a grain boundary or a color variation. The images in the left two columns were produced using the Nikon Stereomicroscope, while the images in the right two columns were produced using the Olympus Inverted Microscope. The yellow rings designate the exact location of the desired landing zones at 1x, 3x, 20x, and 64x magnifications. We used Inkscape (an open-source professional cross-platform vector graphics editor) to process the images for this report.

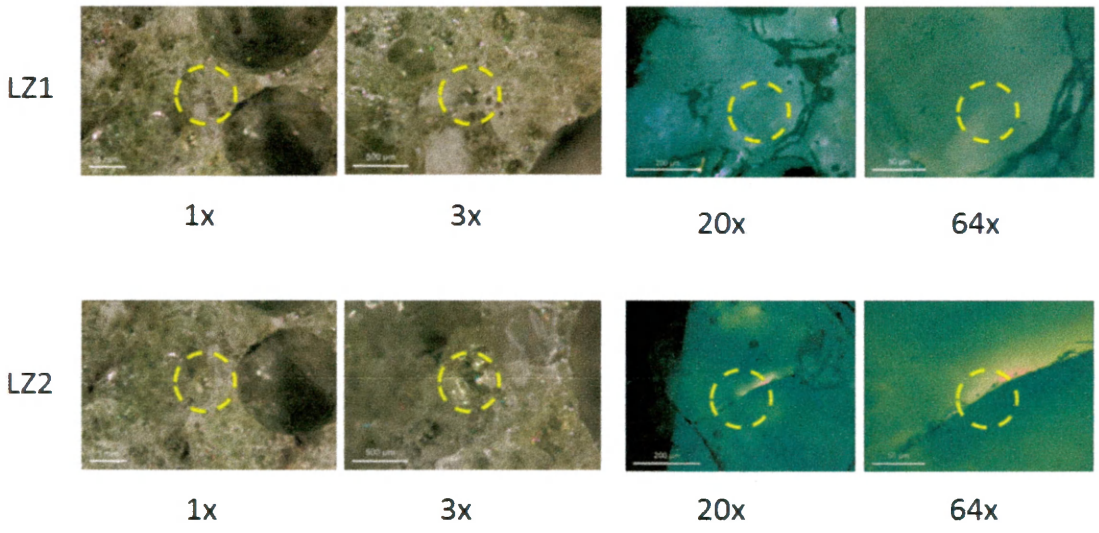


Figure 15: Optical Microscopy - Trinitite Section.

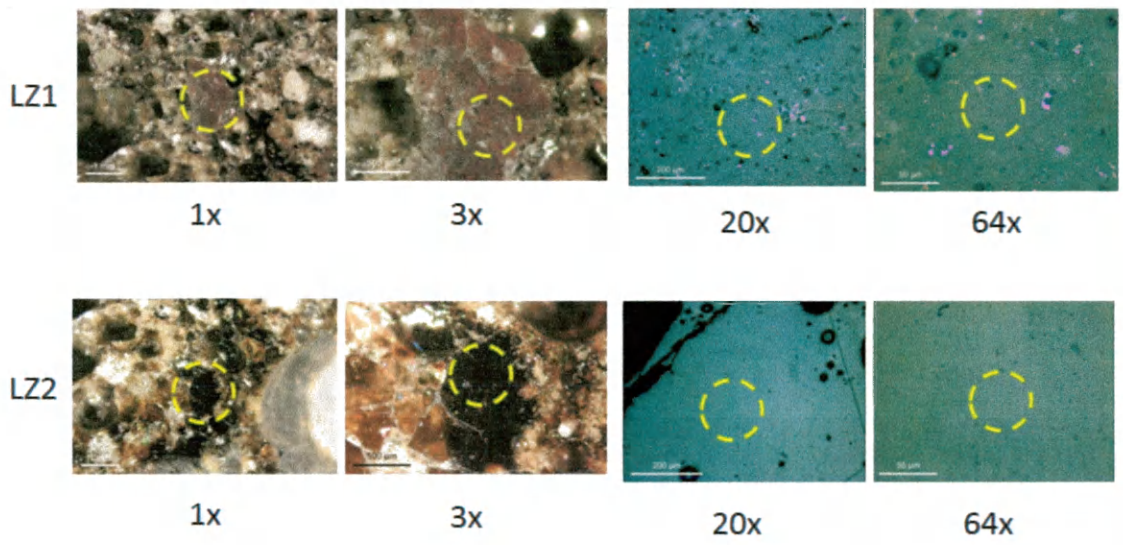


Figure 16: Optical Microscopy - Kharitonchik Section.

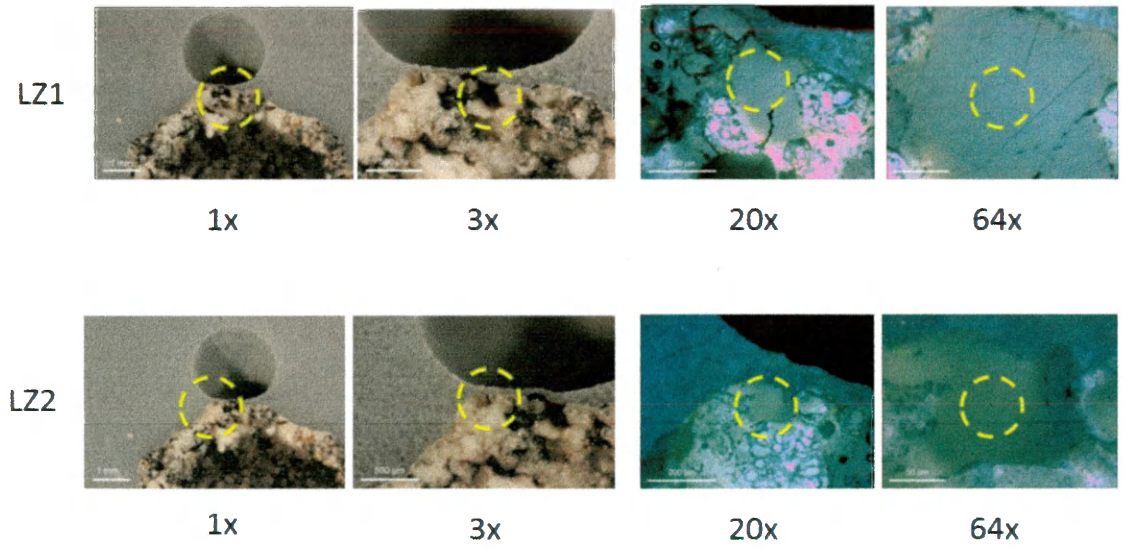


Figure 17: Optical Microscopy - Fulgurite Section.

3.3 Height and Friction Signals

The next six figures (Fig. 18-23) depict AFM images taken over a five micron by five micron area for each sample type (or section) at each of the two pre-designated landing zones (LZ1 and LZ2). The top image shows the height signal scan in nanometers (reflecting the sample's surface topography and morphology). These height signal scans underwent image processing using the NT-MDT NOVA Software's organic image flattening and fitting functions. The bottom image shows the friction signal scan in nanoAmps (reflecting the sample's surface roughness). All of the height signal and friction signal scans were edited using Gwyddion, an open-source professional cross-platform modular program for scanning probe microscopy data visualization and analysis. Lastly, Table 1 below reflects the contact mode AFM scanning parameters used for each of the images displayed in Figures 18-23.

Sample	Pixels	Rate	Gain	Set Point
T - LZ1	512	1.0	0.35	- 1.7
T - LZ2	512	1.0	0.30	- 1.7
K - LZ1	512	0.75	0.75	- 0.1
K - LZ2	512	0.75	0.35	- 0.1
F - LZ1	512	1.0	0.35	- 2.0
F - LZ2	512	1.0	0.40	- 1.8

Table 1: Contact Mode AFM Scanning Parameters

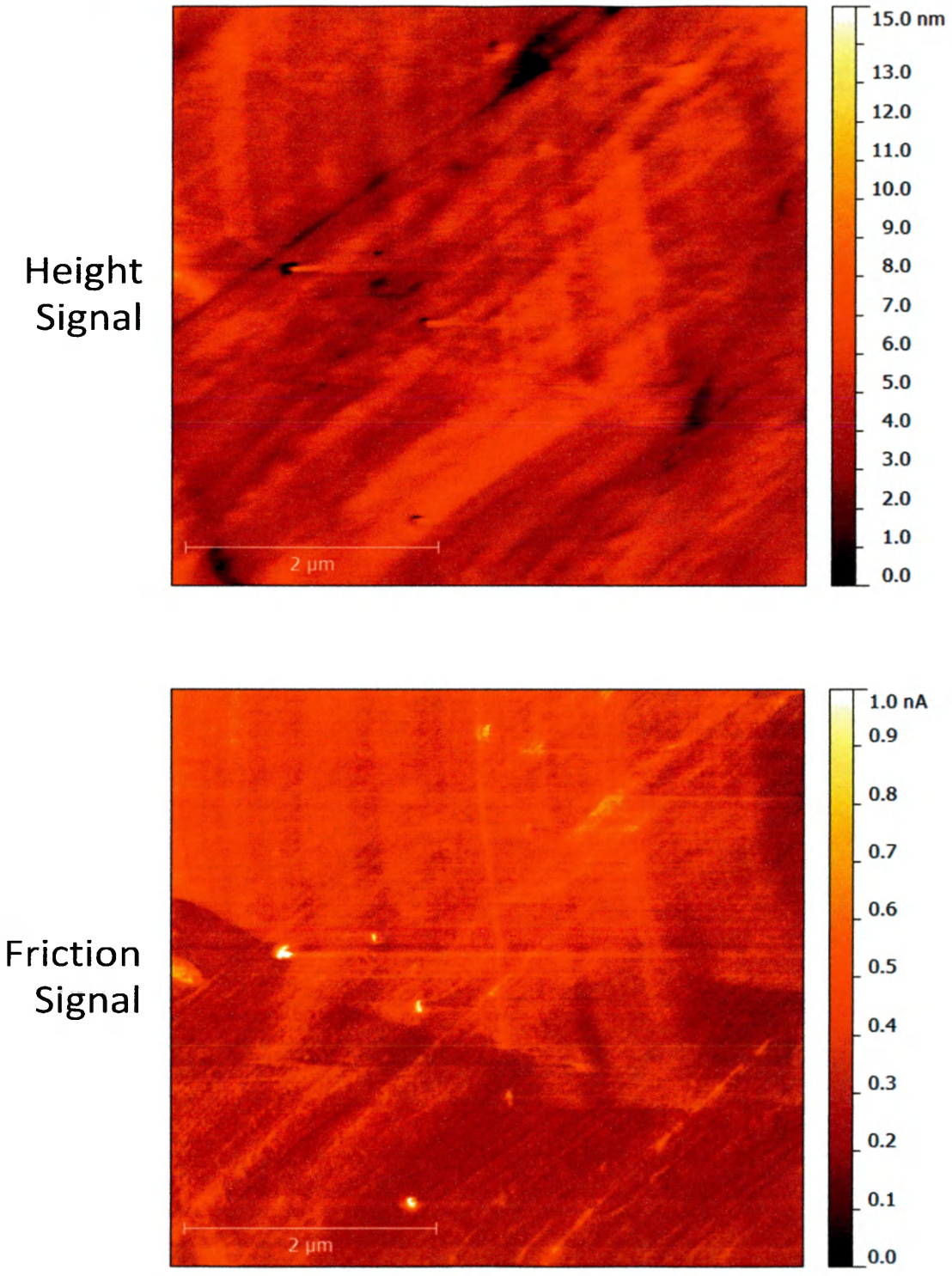


Figure 18: Trinitite LZ1 Height and Friction Signals (5 µm x 5µm scan size).

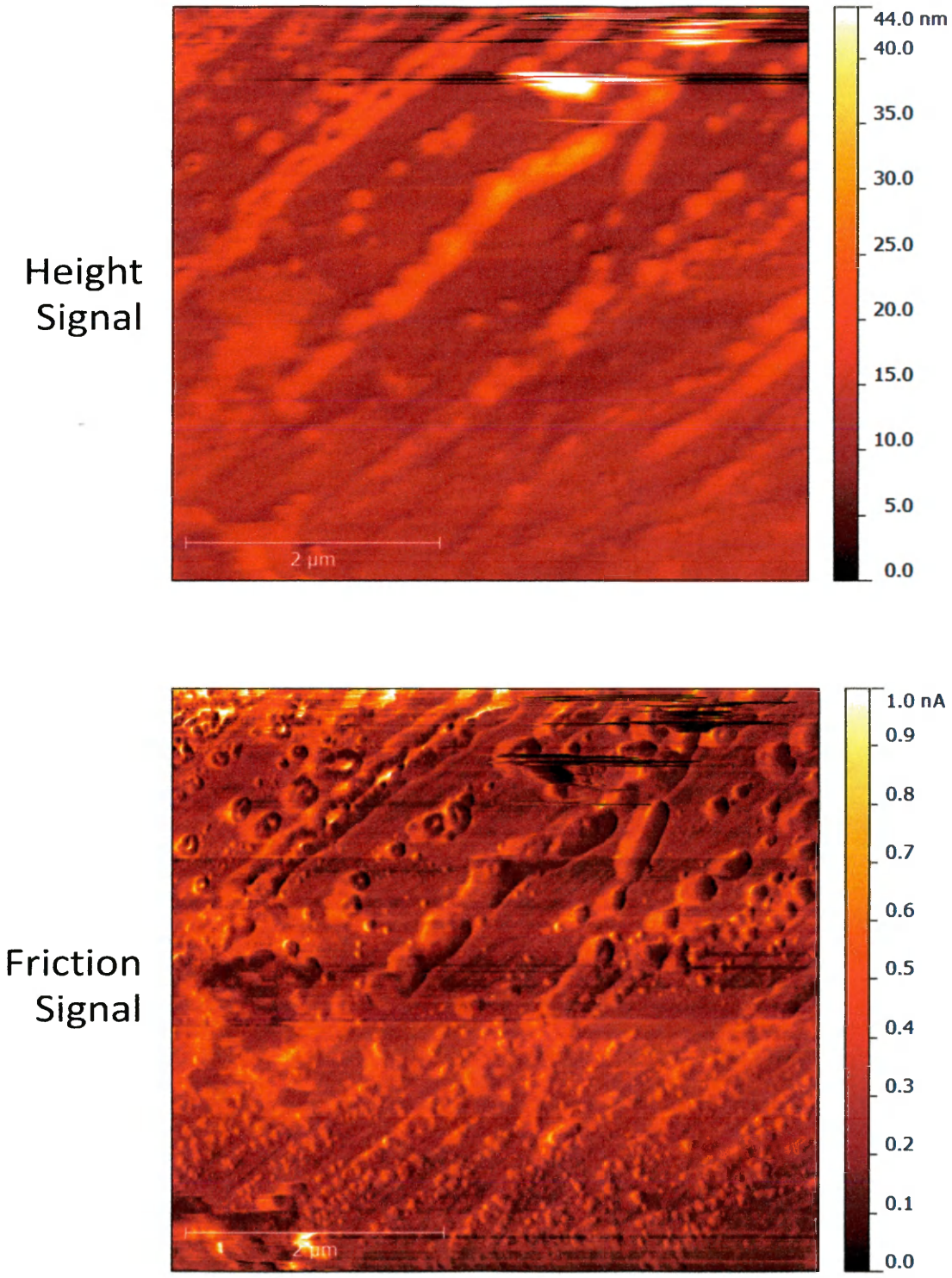


Figure 19: Trinitite LZ2 Height and Friction Signals (5 µm x 5µm scan size).

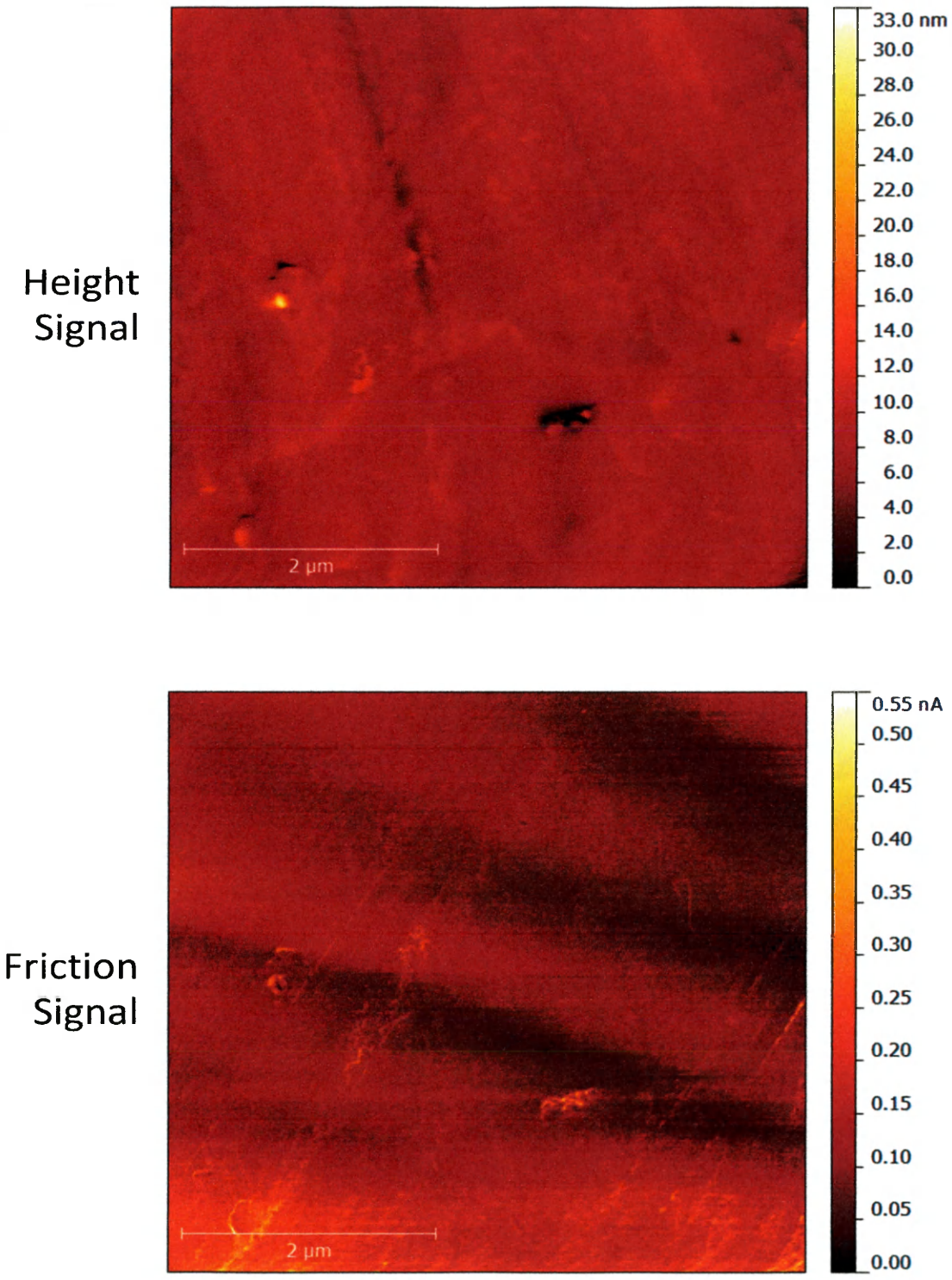


Figure 20: Kharitonchik LZ1 Height and Friction Signals (5 µm x 5µm scan size).

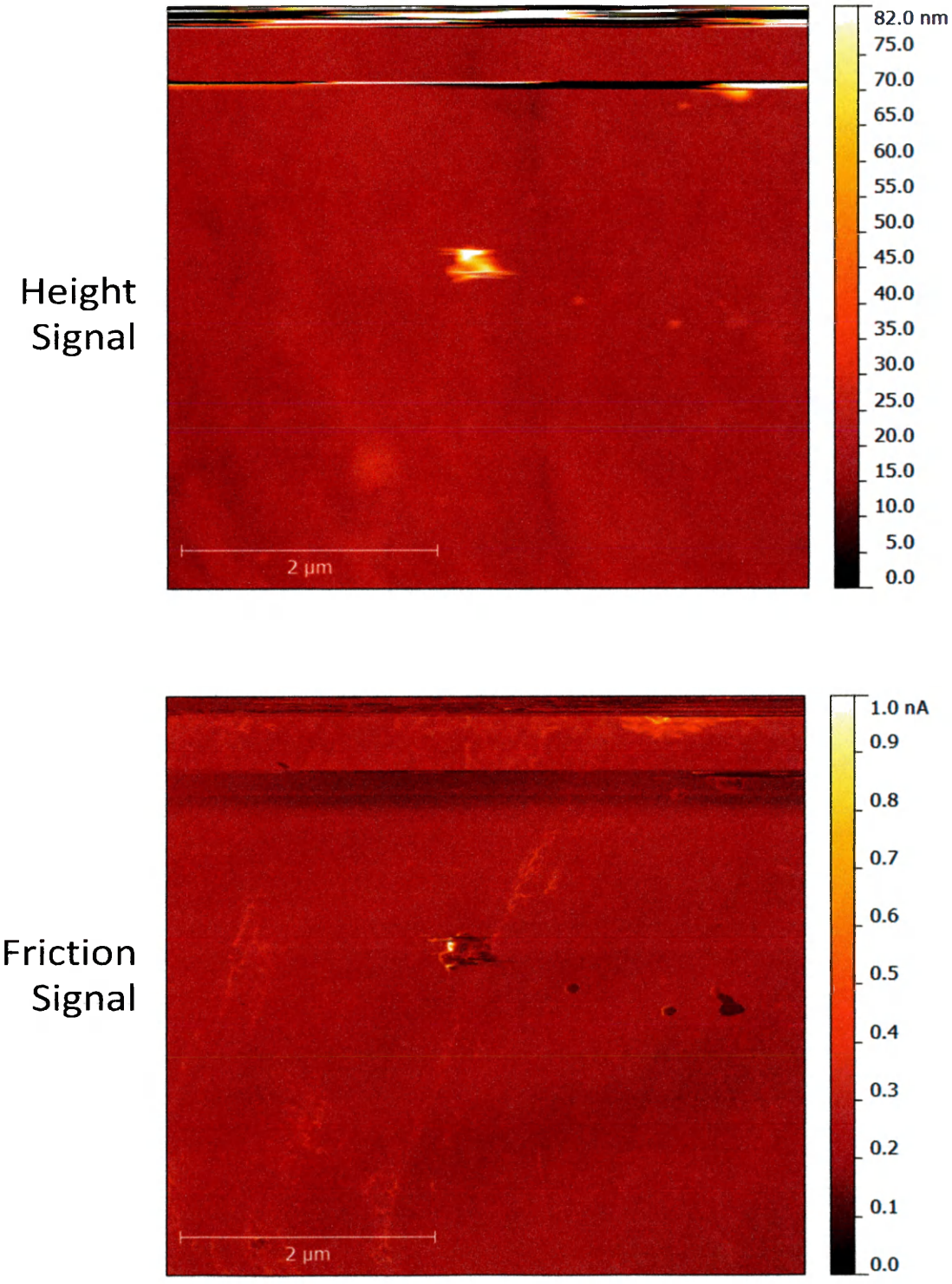


Figure 21: Kharitonchik LZ2 Height and Friction Signals (5 µm x 5µm scan size).

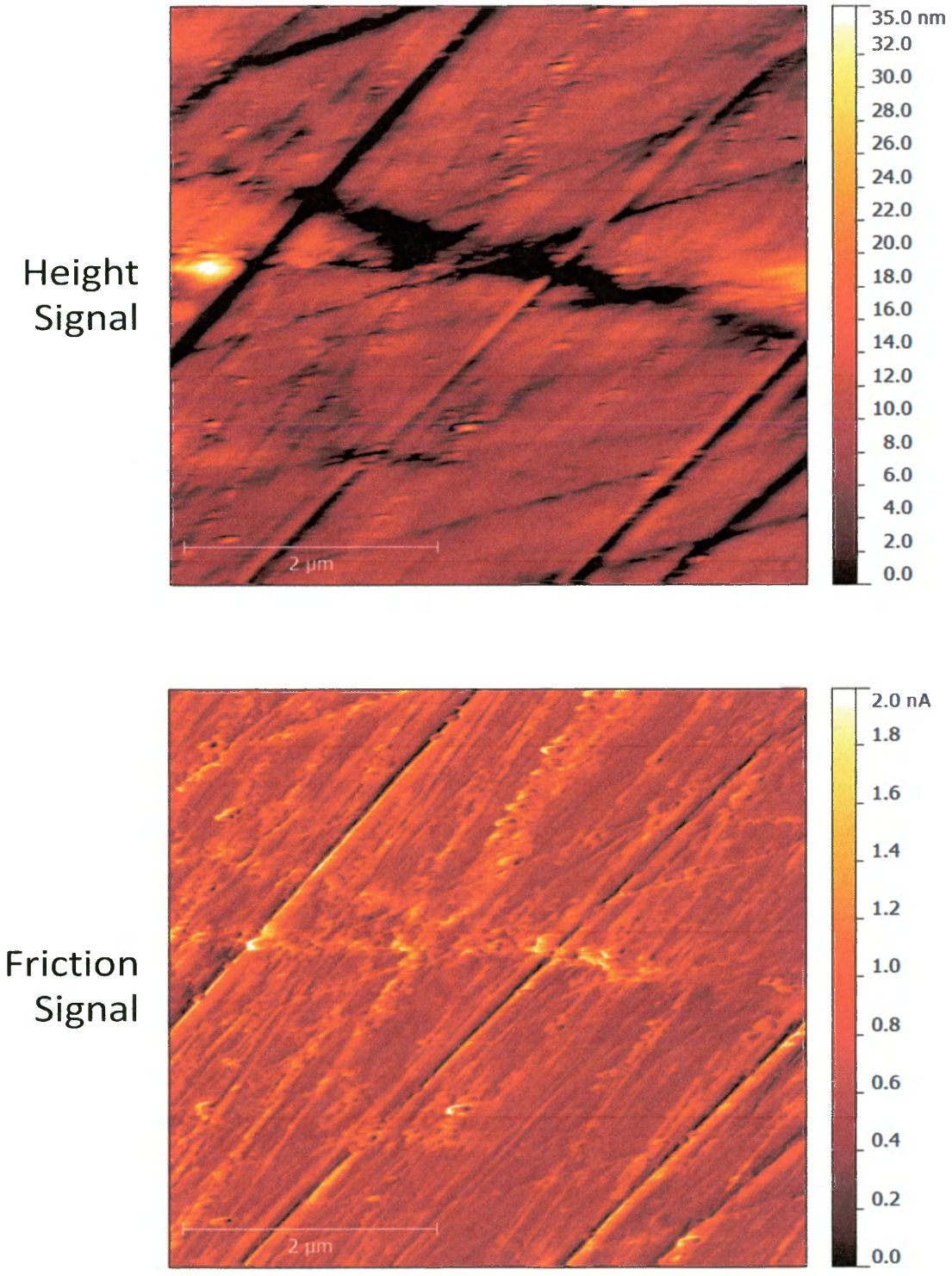


Figure 22: Fulgurite LZ1 Height and Friction Signals (5 µm x 5µm scan size).

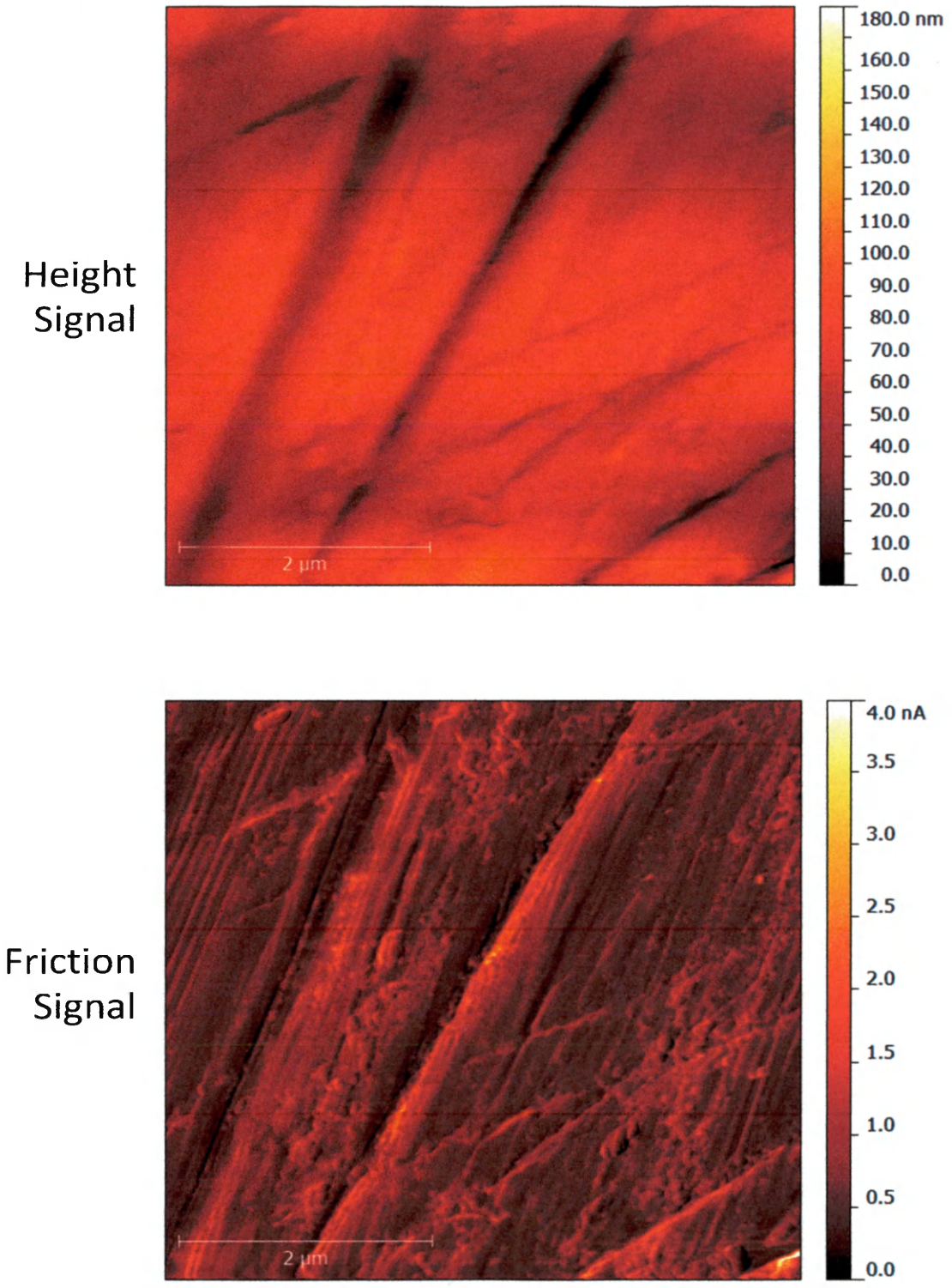


Figure 23: Fulgurite LZ2 Height and Friction Signals (5 µm x 5µm scan size).

Regarding the Trinitite scans (Figures 18 and 19), there is a noticeable difference in the height range between the two pre-designated landing zones on the Trinitite sample. The height range for LZ1 (Figure 18) was only 15 nm, indicating a fairly flat and smooth surface, while the height range for LZ2 (Figure 19) was 44 nm, indicating the higher topographical features associated with the region that appeared to be a grain boundary or a material fracture under optical microscopy. These topographical features or ridges are approximately 0.2 to 0.25 μm wide and approximately 20 to 30 nm high. With regards to the Friction Signals (bottom images), a color variation that was not simply caused by a topographical feature in the Height Signal (top images), indicates a difference in the friction force coefficient and, therefore, a difference in surface chemical composition. This effect is rather pronounced in the Friction Signal image in Figure 18 (LZ1) and may be indicative of the heterogeneous mineraloid nature of the Trinitite sample at that location. Furthermore, the scratches that appear in Figure 18 may have been caused by the sanding and polishing process. Picking up, rotating, and then reapplying the sections onto the rotating disc could account for the fact that the scratches appear in two separate directions. Lastly, the horizontal stripes visible in the Height Signal and Friction Signal on Figure 19 are most likely due to some surface debris picked up by the tip.

Regarding the Kharitonchik scans (Figures 20 and 21), there are noticeable differences between the Friction Signal and the Height Signal for LZ1, however, the banding observed in the Friction Signal image may be the result of

an optical interference pattern caused by the laser scattering off of the sample surface. The use of a high-pass filter on this image may remove this banding effect and enhance the surface roughness variations underneath, which are clearly not associated with any topography on the corresponding height signal image. Furthermore, the height signal image for LZ2 (Figure 21) shows a topographical feature equal to or exceeding 82 nm. Because the rest of the sample surface appears to be uniformly under 45 nm this may be indicative of a surface contaminant or inclusion.

Regarding the Fulgurite scans (Figures 22 and 23), the parallel scratches in these images (which are fairly consistent between the Height and Friction signals) may be due to nano-scale abrasions induced during sample preparation using the Variable Speed Grinder-Polisher at 120 rpm with the 400, 600, 800, and 1200 girt sanding and polishing discs. However, while the scratches appear to be approximately 10.0 to 12.0 nm deep and approximately 0.1 nm wide in Figure 22 and approximately 80.0 to 90.0 nm deep and approximately 0.2 nm wide in Figure 23, these measurements are much smaller than the 6.5 μm grain size associated with the 1200 grit polishing disc, as discussed in section 3.1.

3.4 Three-Dimensional Surface Topography Images

The next seven figures (Fig. 24-30) depict three-dimensional surface topography images, over a five micron by five micron area, for each of the three sample types (or sections), at each of the two pre-designated landing zones. These images graphically represent the morphology, topography, and micro-structural surface geometries at each of the six locations. We used GIMP (GNU Image Manipulation Program...an open-source professional cross-platform raster graphics editor) to prepare the three-dimensional surface topography images for this report. Lastly, Figure 30 displays all six of the three-dimensional surface topography images together for a qualitative comparison.

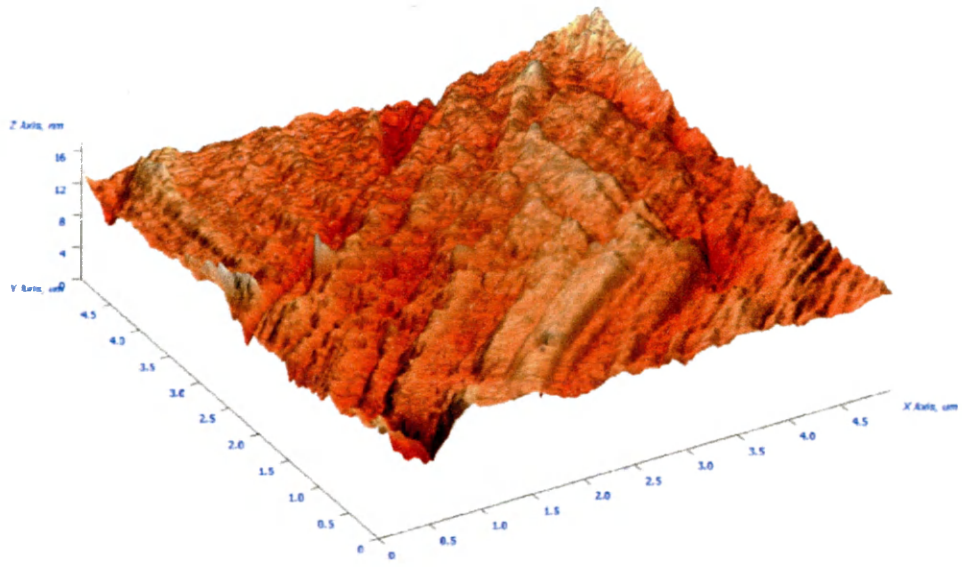


Figure 24: Trinitite LZ1 3D Surface Topography Image.

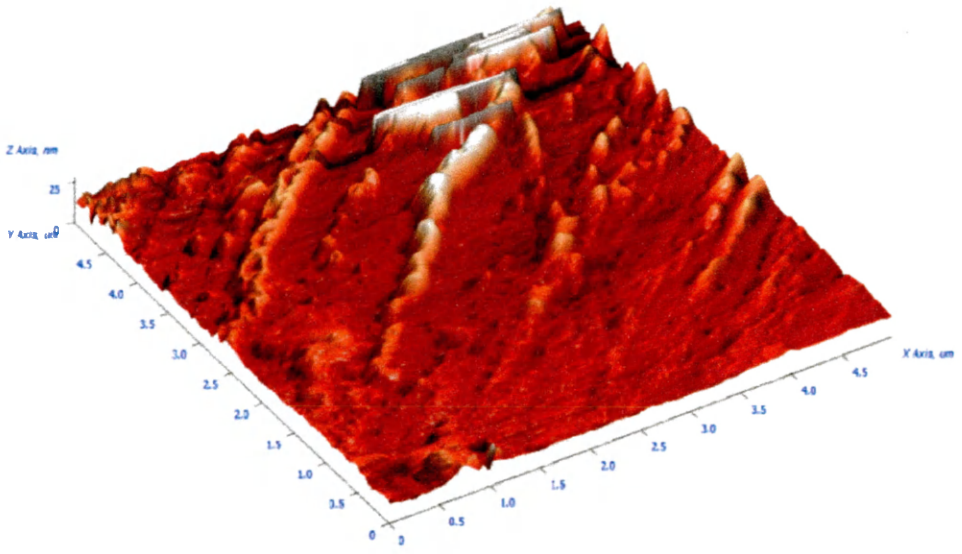


Figure 25: Trinitite LZ2 3D Surface Topography Image.

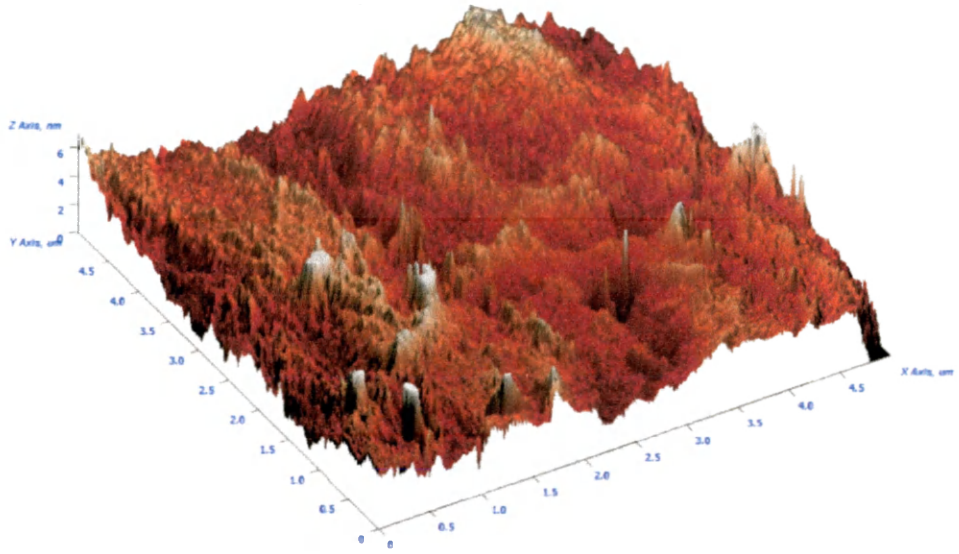


Figure 26: Kharitonchik LZ1 3D Surface Topography Image.

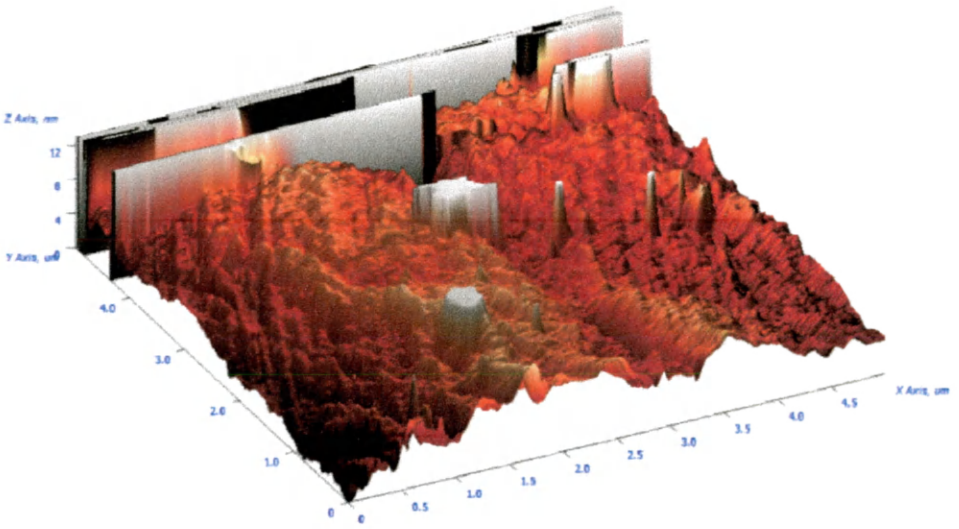


Figure 27: Kharitonchik LZ2 3D Surface Topography Image.

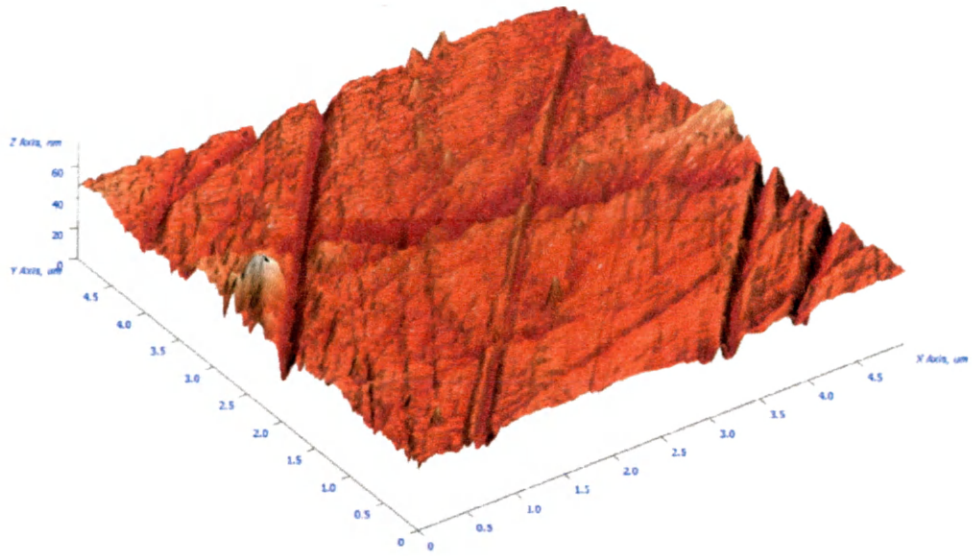


Figure 28: Fulgurite LZ1 3D Surface Topography Image.

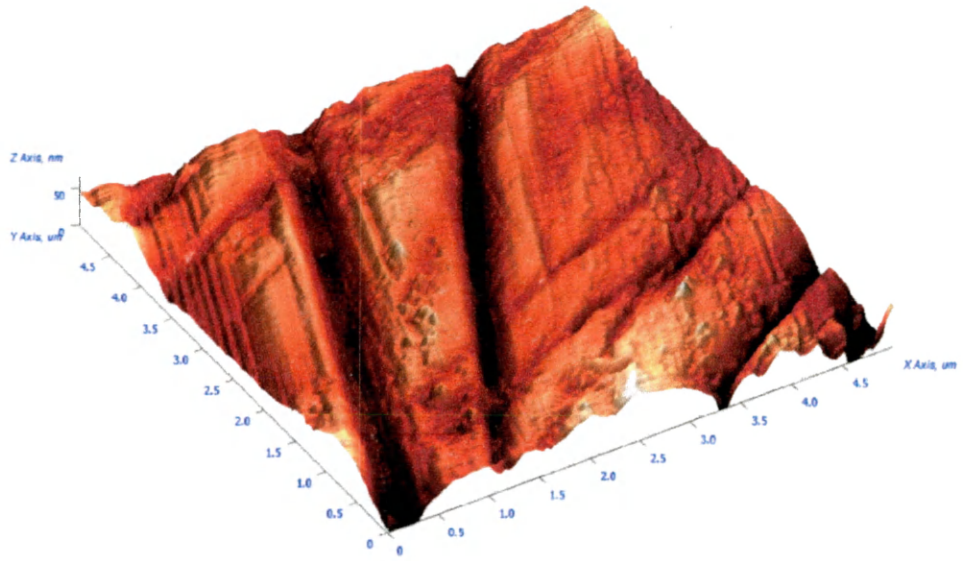


Figure 29: Fulgurite LZ2 3D Surface Topography Image.

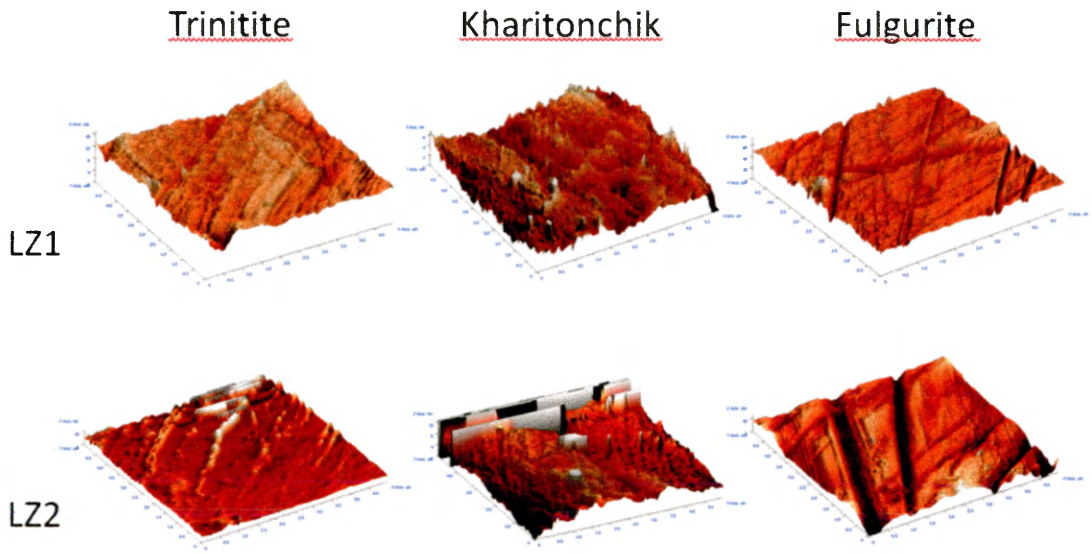


Figure 30: 3D Surface Topography Images Combined for Comparison.

3.5 Force Spectroscopy (Force Curves)

The next seven figures (Fig. 31-37) depict the force curves taken for each of the three sample types (or sections), at each of the two pre-designated landing zones, on the center point of each five micron by five micron scan. These force curves reflect the surface adhesion at that particular point on the surface. The x-axis represents the piezo displacement in nanometers, while the y-axis represents the cantilever deflection in nanoAmps. The red curve traces the tip approach, while the blue curve traces tip retract. Figure 37 displays all six of the force curves together for a qualitative comparison. Lastly, Table 2 below reflects the relevant input parameters for each of the force curves displayed in Figures 31-37.

Sample	Mode	Piezo Displacement Range	Points	Cantilever Deflection Range
T - LZ1	DFL (2)	4000 to - 250.0 nm	2000	- 50.0 to 4.0 nA
T - LZ2	DFL (2)	3000 to - 250.0 nm	2000	- 50.0 to 4.0 nA
K - LZ1	DFL (2)	700.0 to 0.0 nm	2000	- 50.0 to 2.0 nA
K - LZ2	DFL (2)	700.0 to 0.0 nm	2000	- 50.0 to 2.0 nA
F - LZ1	DFL (2)	6000 to 0.0 nm	2000	- 50.0 to 4.0 nA
F - LZ2	DFL (2)	6000 to 0.0 nm	2000	- 50.0 to 4.0 nA

Table 2: Force Curve Input Parameters.

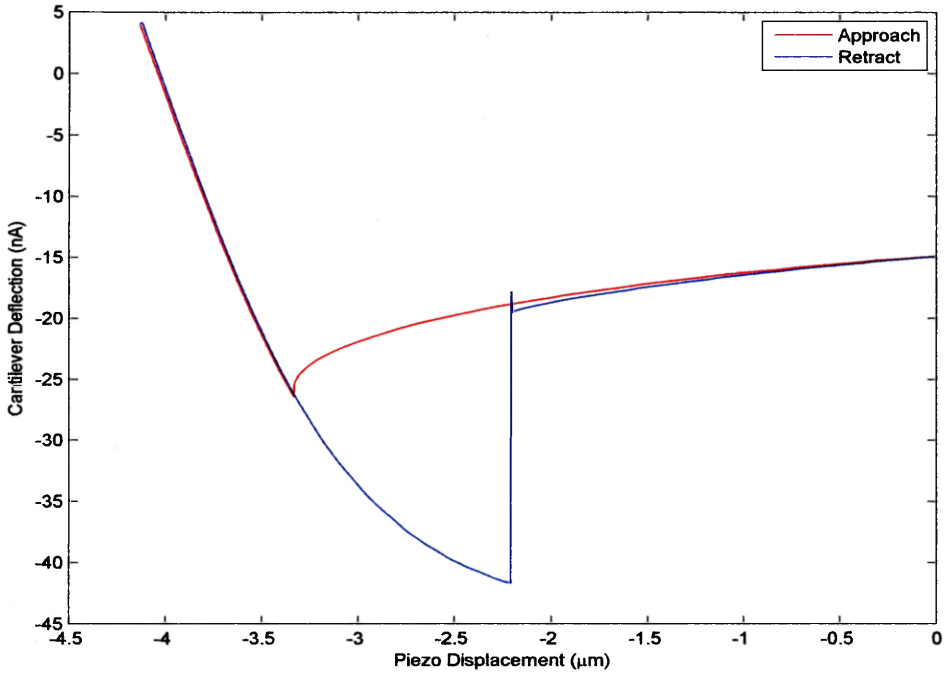


Figure 31: Trinitite LZ1 Force Curve.

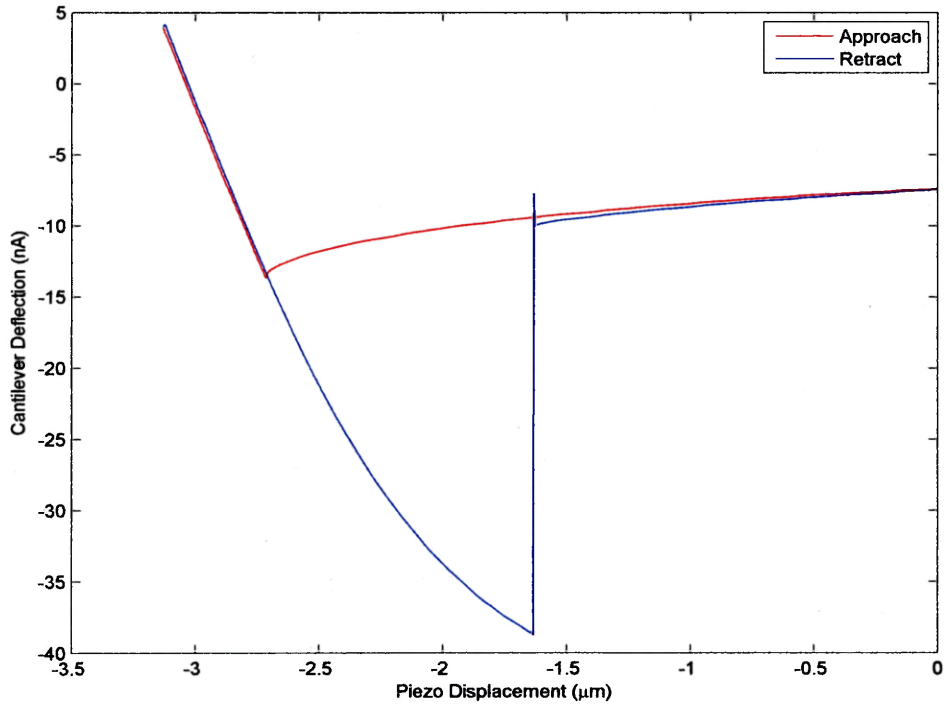


Figure 32: Trinitite LZ2 Force Curve.

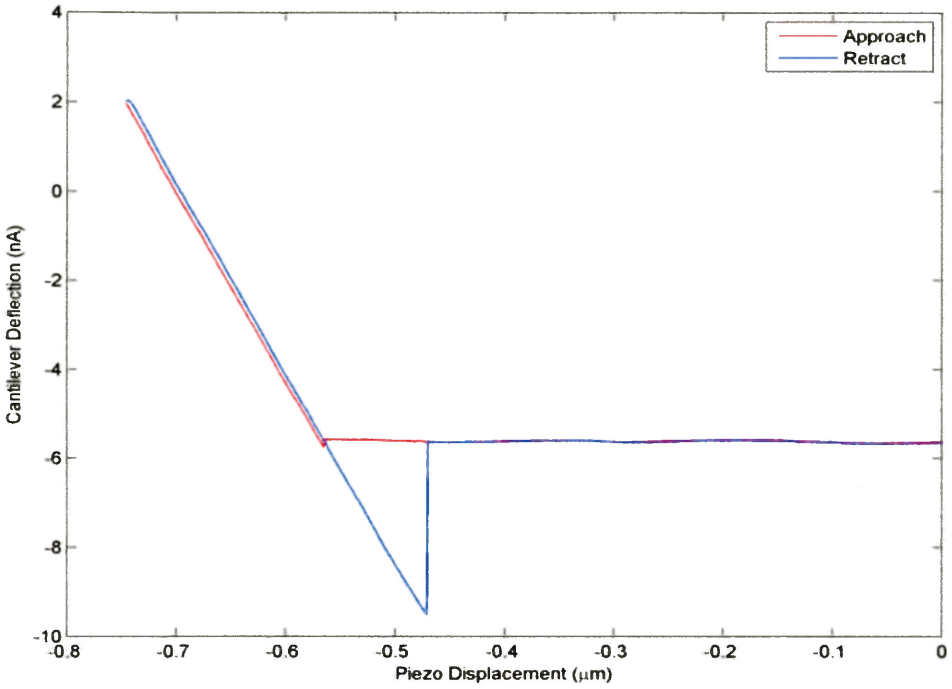


Figure 33: Kharitonchik LZ1 Force Curve.

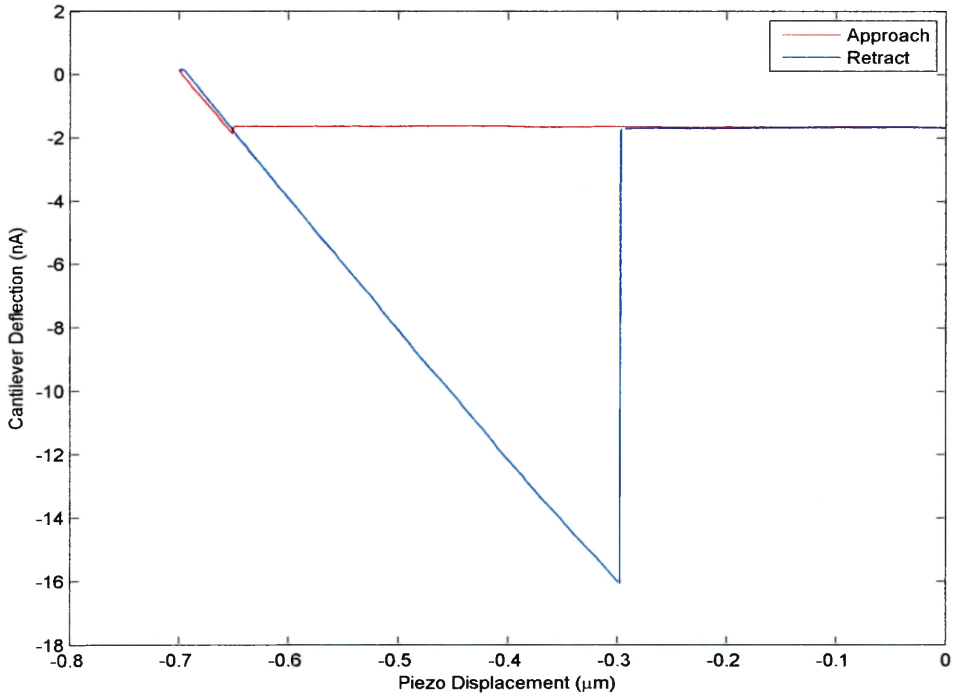


Figure 34: Kharitonchik LZ2 Force Curve.

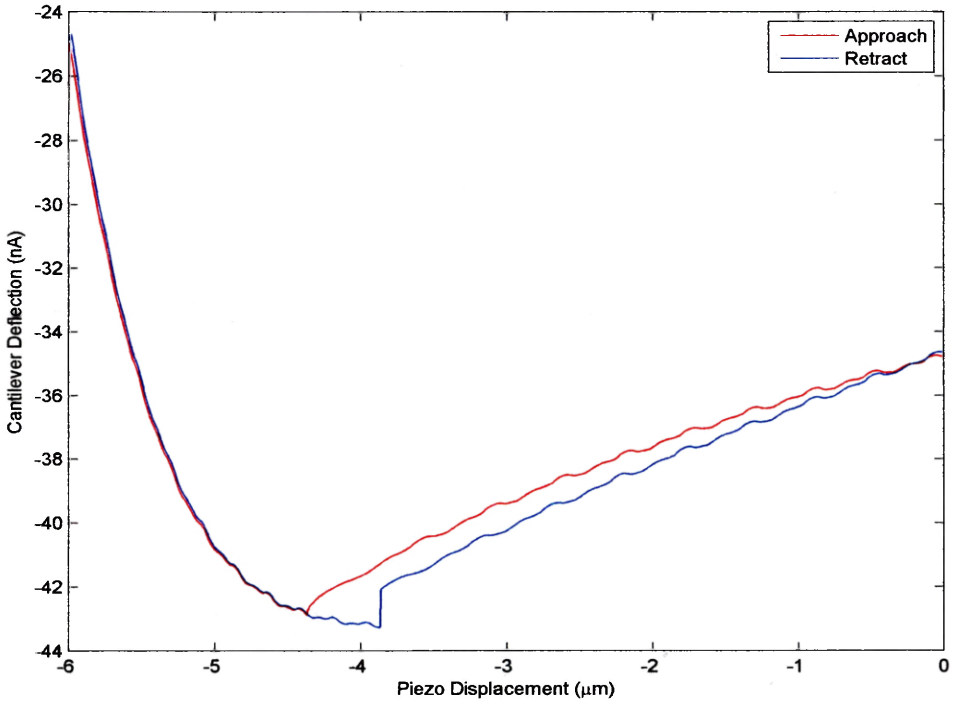


Figure 35: Fulgurite LZ1 Force Curve.

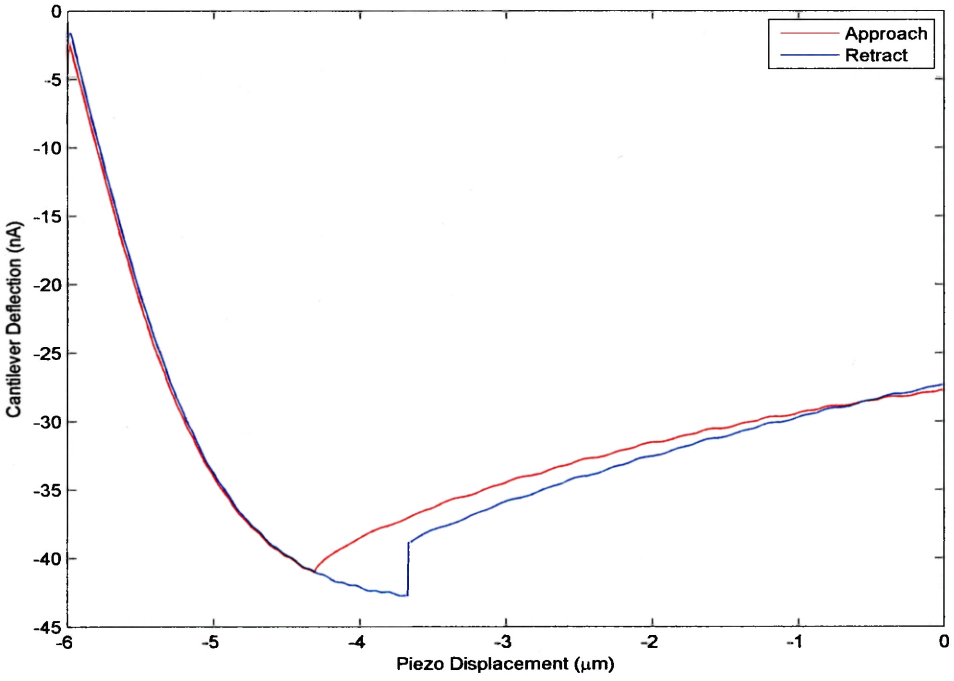


Figure 36: Fulgurite LZ2 Force Curve.

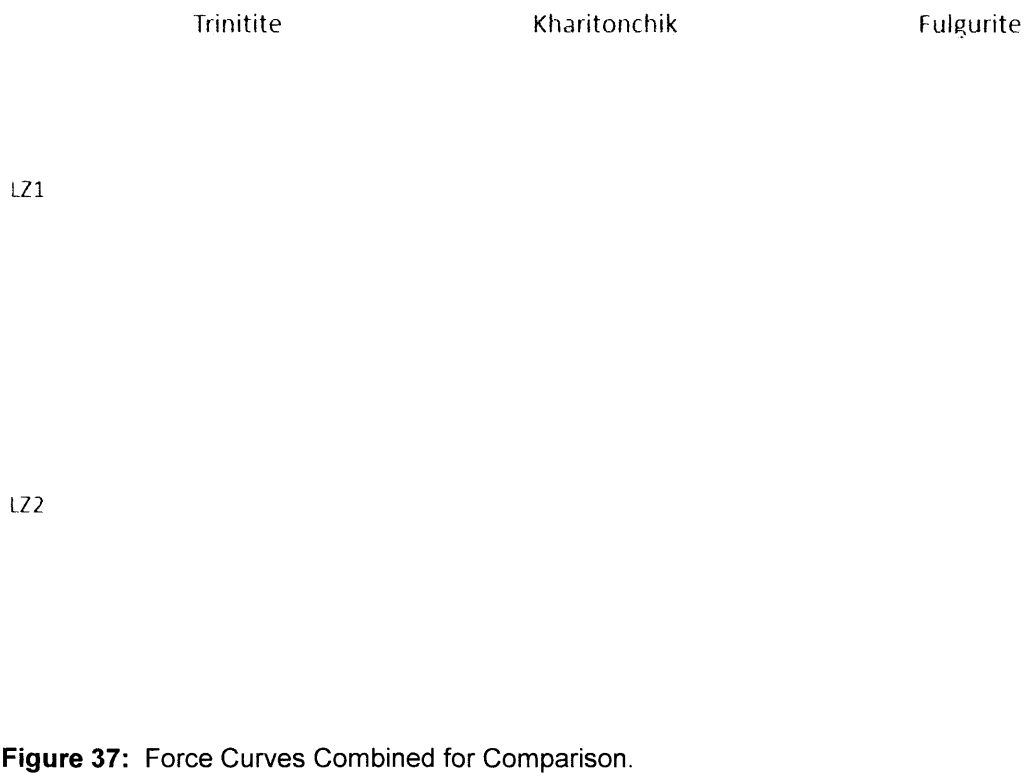


Figure 37: Force Curves Combined for Comparison.

The Kharitonchik Force Curves demonstrate a fairly classic relationship and a linear appearance. On the other hand, the Force Curves produced by the Trinitite and Fulgurite samples are curved, potentially demonstrating a "non-linear optical lever sensitivity" (Thormann, 2009) due to what we speculate are strongly attractive electrostatic Coulomb forces between the tip and the sample surface for those sample types (or sections). In simple terms, the attractive forces were so strong that the negative laser deflection exceeded the $\pm$ 30 nA photo diode range, resulting in the curvature of the plots. Thormann et al. (2009) concluded that it is difficult to correctly "convert detector output into real cantilever deflection" when additional surface forces, like long-range electrostatic forces, are experienced. Regardless, by examining the required "pull-off" distance

versus the difference in electric current produced by the cantilever deflection for each Force Curve (the adhesive regime depicted as region "D" in Fig. 6), one can qualitatively deduce that we were able to detect different adhesive forces between the different sample types (or sections) and even between different locations (LZs) on the same samples.

4.0 DISCUSSION

The key to Technical Nuclear Forensics is to be able to identify forensically useful markers or signatures (like specific impurities, crystal structures, surface finishes, contaminants, alloying or cladding material) linking the perpetrators to specific factors such as locations, events, materials, processes, time, weapon type, and so on (Joint Working Group, 2008). With regards to the analysis of post-detonation materials and debris, these investigations might provide insight into the fuel type, fuel amount, physical makeup, operational efficiency, and design sophistication of the device. With regards to the analysis of pre-detonation materials, these investigations might provide insight into a particular nuclear material's production cycle (mining, refining, enrichment), use in reactors, recycling, or intended purpose. This initial research focuses primarily on the analysis of post-detonation materials. The reason that this research is so novel is that, while previous technical nuclear forensic research and studies involve the use of a variety of characterization tools and techniques, there is almost no mention of AFM in the available literature. At the unclassified level, this author is aware of only one other research group (at the Argon National Laboratory) that is investigating the use of AFM to conduct Technical Nuclear Forensics on radioactive materials. In fact, this research group just received a Domestic Nuclear Detection Office research grant in 2013 under the Nuclear Forensics Expertise Development Program.

With regards to the specific results we obtained using contact mode AFM to produce the Height and Friction Signal scans (Fig. 18-23), it is important to note that the natural surfaces of these melt-glass samples were particularly rough and uneven. Sanding and polishing the samples, while altering some of the natural surface features, provided many benefits, not least, of which, was the ability to land the tip and scan the material. In addition, it improved our ability to acquire lateral friction force scans. Furthermore, it was envisioned that these sanded and polished sample sections would be particularly well-suited for subsequent research involving dynamic, semi-contact (tapping-mode) or non-contact mode AFM imaging. Lastly, each of the Height Signal scans underwent additional image processing to correct for substrate tilt by subtracting the surface (using the 2D Flatten Correction Plane Fit) and to correct for unwanted artifacts by fitting the lines (using the 1D Flatten Correction First Order Fit). In some cases, a Histogram Fit was performed, as well.

With regards to the three-dimensional Surface Topography Images (Fig. 24-30), these nano-scale graphical representations enhance our qualitative understanding of the sample surface at these locations, however, further quantitative analysis techniques, for instance measuring "maximum peak heights (Hpm), average maximum height (Hz), average maximum valley depths (Hvm), peak-to-valley distances (Rz), and the root mean square roughness (Rms)" using image analysis software (like Image J) would be the logical next step (Chen, 2011). These kinds of surface morphology parameters would provide more

precise and measurable information regarding pore shape and size, grain shape and size, grain boundary morphology, and micro-structural orientation. This information may, in turn, provide valuable insight regarding parameters like the heat, pressure, fuel, efficiency, and height of burst. In cases involving pre-detonation materials, this kind of quantitative analysis could provide valuable insight regarding, for example, specific signatures and markers associated with uranium oxide pellet fabrication to include particle size distribution and particle inclusions or occlusions (IAEA, 2006).

With regards to Force Spectroscopy or taking Force Curves (Fig. 31-37), there are a few important things to understand. Unlike dynamic mode AFM, the cantilever is not "driven" (vibrated) and the X & Y directions of the piezo are fixed, while the piezo moves only in the Z direction in order to approach the cantilever to the surface and retract it back to the initial position, measuring the tip-surface potential (stiffness and adhesion), in accordance with the Lennard-Jones Potential discussed previously (Fig. 3). These Force Curves are useful as a qualitative characterization of the tip-surface potential and not a quantitative analysis. To perform a quantitative analysis, one would have to determine the actual spring constant of the cantilever and then calculate the deflection calibration of that particular cantilever. Although challenging, it is certainly not impossible to do, however, it exceeded the scope of this initial research project.

After working with these materials for a few months, experimenting with different sample preparation techniques, and employing various AFM modes, we

successfully realized many of the initial research goals and objectives. However, we also observed that the characterization and analysis of post-detonation material requires a fair degree of reverse-engineering, which can be rather challenging. Therefore, we speculate that the analysis of "known" pre-detonation materials for specific properties may be more straight forward and thus present a rather lucrative target for additional AFM analysis. In fact, it is presumed that "nuclear forensics can, with high reliability, reach certain conclusions but those may not be sufficient to uniquely identify the source. The more extensive the database and libraries of sample materials and associated isotopic analyses are, the more specific attribution can be" (Joint Working Group, 2008).

Perhaps, in the future, we can expand on our initial qualitative force spectroscopy work to include quantitative analysis techniques. We can explore AFM dynamic modes to include semi-contact and non-contact scanning to analyze surface energy and stiffness. We can investigate the use of specialized or modified AFM probes to detect specific markers or signatures like chemical impurities, surface contaminants, and material inclusions. We can attempt to navigate the requirements for conducting classified research and incorporate investigations using radioactive samples and pre-cursor materials. And lastly, we can consider using other characterization tools like Scanning Electron Microscopy and Petrography, together with AFM, in order to improve our overall understanding of the samples under investigation.

5.0 CONCLUSION

In conclusion, we explored a novel approach to Technical Nuclear Forensics utilizing Atomic Force Microscopy as part of the counter-response to a very serious problem...a terrorist attack using a nuclear or radiological device. We conducted an initial proof of concept, using qualitative analysis to perform a comparative nano-scale characterization of three specifically selected melt-glass samples representing post-detonation materials. And we proposed numerous opportunities for future research in this field.

More specifically, we utilized contact mode AFM to successfully collect very precise three-dimensional height data with nano-scale resolution from samples of post-detonation material reflecting surface morphology and topography. In addition, we obtained lateral friction force data that graphically demonstrated variations in surface roughness (or friction coefficients) indicating the presence of different chemical compositions at the surface of these materials. Furthermore, we produced force curves that qualitatively demonstrate variations in surface adhesion between the various melt-glass samples, as well as, between various locations on the same melt-glass sample. And lastly, we were able to detect a significant difference in electrostatic coulomb forces between various sample types, which was an unexpected but very interesting outcome meriting further investigation.

Some potential applications of this initial investigation and subsequent research efforts include supporting various non-proliferation initiatives such as the Domestic Nuclear Detection Office-led interagency effort to develop a Global Nuclear Detection Architecture, supporting the development of the International Atomic Energy Agency's Strengthened Safeguards System, and supporting the known nuclear explosive materials international database. Furthermore, this type of research could lead to direct support of the National Technical Nuclear Forensics mission by developing and fielding a hardened and deployable AFM-type device for response operations. With some modification, particularly with respect to working with hot radioactive sources, a system leveraging all the advantages listed in section 1.2 could prove to be useful in operational settings. However, more research would have to be conducted in order to determine the feasibility of this approach.

In closing, we determined that AFM may be valuable to the technical nuclear forensics mission, particularly, if used in conjunction with other analytical methods and characterization tools. Further research using additional AFM modes and quantitative analysis techniques is warranted and may lead to further collaboration with NASA Langley Research Center, Argon National Laboratory, and the Defense Threat Reduction Agency, as well as, future consideration under the Domestic Nuclear Detection Office Nuclear Forensics Expertise Development Program.

BIBLIOGRAPHY

Beasley, T. M., Kelley, J., Orlandini, K., Bond, L., & Aarkrog, A. (1998). Isotopic pu, U, and np signatures in soils from semipalatinsk-21, kazakh republic and the southern urals, russia. *Journal of Environmental Radioactivity, 39*(2), 215; 215-230; 230.

Belloni, F., Himbert, J., Marzocchi, O., & Romanello, V. (2011). Investigating incorporation and distribution of radionuclides in trinitite. *Journal of Environmental Radioactivity, 102*(9), 852-862. doi:10.1016/j.jenvrad.2011.05.003

Bellucci, J. J., Simonetti, A., Wallace, C., Koeman, E. C., & Burns, P. C. (2013). Isotopic fingerprinting of the world's first nuclear device using post-detonation materials. *Analytical Chemistry, 85*(8), 4195-4198. doi:10.1021/ac400577p; 10.1021/ac400577p

Bellucci, J. J., Simonetti, A., Wallace, C., Koeman, E. C., & Burns, P. C. (2013). Lead isotopic composition of trinitite melt glass: Evidence for the presence of canadian industrial lead in the first atomic weapon test. *Analytical Chemistry, 85*(15), 7588-7593. doi:10.1021/ac4016648; 10.1021/ac4016648

Bellucci, J. J., & Simonetti, A. (2012). Nuclear forensics: Searching for nuclear device debris in trinitite-hosted inclusions. *Journal of Radioanalytical and Nuclear Chemistry, 293*(1), 313-319. doi:10.1007/s10967-012-1654-9

Bellucci, J. J., Wallace, C., Koeman, E. C., Simonetti, A., Burns, P. C., Kieser, J., & Walczak, T. (2013). Distribution and behavior of some radionuclides associated with the trinity nuclear test. *Journal of Radioanalytical and Nuclear Chemistry, 295*(3), 2049-2057. doi:10.1007/s10967-012-2201-4

Betti, M., Tamborini, G., & Koch, L. (1999). Use of secondary ion mass spectrometry in nuclear forensic analysis for the characterization of plutonium and highly enriched uranium particles. *Analytical Chemistry, 71*(14), 2616-2622. doi:10.1021/ac981184r

Cetron, M. J., & Davies, O. (2008). 55 trends now shaping the future of terrorism. *National Intelligence University, Office of the Director of National Intelligence and the Center for Strategic Leadership, U.S. Army War College Proteus Trends Series* Vol 1, Issue 2, 1-254.

Chen, Y. (2011). Forensic applications of nanotechnology. *Journal of the Chinese Chemical Society*, 58, 828-835

Evseeva, T., Belykh, E., Geras'kin, S., & Majstrenko, T. (2012). Estimation of radioactive contamination of soils from the "Balapan" and the "Experimental field" technical areas of the semipalatinsk nuclear test site. *Journal of Environmental Radioactivity, 109*(0), 52-59. doi:http://dx.doi.org/10.1016/j.jenvrad.2012.01.002

Eaton, P., & West, P. (2010). Atomic Force Microscopy. New York, NY: Oxford University Press.

Esce, J. D. (2013). NTNF briefing slides. Retrieved from 20th CBRNE, G37, Exercise Section

Fahey, A. J., Ritchie, N. W. M., Newbury, D. E., & Small, J. A. (2010). The use of lead isotopic abundances in trace uranium samples for nuclear forensics analysis. *Journal of Radioanalytical and Nuclear Chemistry, 284*(3), 575-581. doi:10.1007/s10967-010-0509-5

Fahey, A. J., Zeissler, C. J., Newbury, D. E., Davis, J., & Lindstrom, R. M. (2010). Postdetonation nuclear debris for attribution. *Proceedings of the National Academy of Sciences of the United States of America, 107*(47), 20207-20212. doi:10.1073/pnas.1010631107

Friction Force Graphic. (2013, July 11). Atomic force microscopy. *The Zen of Stem*. Retrieved from http://zenofstem.com/project/atomic-force-microscopy/

Fulgurite. (n.d.). Retrieved March 20, 2013, from Wikipedia, http://en.wikipedia.org/wiki/Fulgurite

Glasser, A., & von Hippel, F., (2006). Thwarting nuclear terrorism: many civilian research reactors contain highly enriched uranium that terrorists could use to build nuclear bombs. *Scientific American*, 294, 56 - 63 doi:10.1038/scientificamerican0206-56

Glasstone, S., & Dolan, P. J. (1977). The Effects of Nuclear Weapons, Third Edition. Washington, DC: U.S. Government Printing Office.

Hastings, E. P., Lewis, C., FitzPatrick, J., Rademacher, D., & Tandon, L. (2008). Characterization of depleted uranium oxides fabricated using different processing methods. *Journal of Radioanalytical and Nuclear Chemistry, 276*(2), 475-481. doi:10.1007/s10967-008-0529-6

Haugstad, G. (2012): Atomic Force Microscopy: Understanding Basic Modes and Advanced Applications. Hoboken, NJ: John Wiley & Sons, Inc.

Hirsh, M. (2013). A glimpse at the next bin laden?. *National Journal Magazine.* Retrieved from http://www.theatlantic.com/international/archive/2013/11/the-next-bin-laden/281538/

International Atomic Energy Agency (IAEA). (2006). Nuclear forensics support reference manual (IAEA nuclear security series, ISSN 1816-9317; no. 2). Vienna, Austria: IAEA.

Jockey's Ridge State Park. (n.d.). In Images for Jockey's Ridge State Park. Retrieved from https://www.google.com/search?q=jockey's+ridge+state+park&tbm=isch&tbo=u&source=univ&sa=X&ei=V25JU_vPI8Xb8gHgloGwAQ&sqi=2&ved=0CFQQsAQ&biw=1366&bih=625

Joint Working Group of the American Physical Society and the American Association for the Advancement of Science. (2008). Nuclear forensics: role, state of the art, program needs (AAAS Center for Science Technology and Security Policy). Washington, DC: U.S. Government Printing Office.

Kentis, S. E., & Ulicny, W. D. (2009). National nuclear forensics expertise development program. *Current Status, Trends, and Needs in Radiochemical Education: The Us and Abroad, 1164*, 47-51.

Kristo, M. J., & Tumey, S. J. (2013). The state of nuclear forensics. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 294*(0), 656-661. doi:10.1016/j.nimb.2012.07.047

Maurer, D.C., & Aloise, G. (2011). Last line of defense: federal, state, and local efforts to prevent nuclear and radiological terrorism within the United States. *U.S. House of Representatives, Committee on Homeland Security, Subcommittee on Cybersecurity, Infrastructure Protection, and Security Technologies*. (DHHS Digital Library, Naval Postgraduate School, Center for Homeland Defense and Security). Washington, DC: U.S. Government Printing Office.

Mayer, K., Wallenius, M., & Fanghänel, T. (2007). Nuclear forensic science—From cradle to maturity. *Journal of Alloys and Compounds, 444–445*(0), 50-56. doi:10.1016/j.jallcom.2007.01.164

Mercer, D. J., Hypes, P. A., & Saey, P. R. (2010). Recent onsite gamma measurements at the Trinity Test Site and a comparison to Trinitite samples. Proceedings of the 52nd Annual Meeting of the Institute for Nuclear

Materials Management, Los Alamos National Laboratory, Los Alamos, NM, LA-UR-11-03597

National Technical Nuclear Forensics Center (NTNFC). (n.d.). National Technical Nuclear Forensics Center Brochure. Retrieved November 13, 2013, from 20th CBRNE, G37, Exercise Section

Office of the Deputy Assistant to the Secretary of Defense (Nuclear Matters). (2008). Nuclear Matters: A Practical Guide (Pentagon). Washington, DC: U.S. Government Printing Office.

Parekh, P., Semkow, T., Torres, M., Haines, D., Cooper, J., Rosenberg, P., & Kitto, M. (2006). Radioactivity in trinitite six decades later. *Journal of Environmental Radioactivity, 85*(1), 103-120. doi:10.1016/j.jenvraf.2005.01.017

Pasek, M., Block, K., & Pasek, K. (2012). Fulgurite morphology: a classification scheme and clues to formation. Contrib Mineral Petrol 164:477-492 doi: 10.1007/s00410-012-0753-5

Phillips, M. (2007). Uncertain justice for nuclear terror: deterrence of anonymous attacks through attribution. *Foreign Policy Research Institute*, Elsevier Limited 429-446.

Porter, R., Ruggiero, C., Hush, D., Harvey, N., Kelly, P., Scoggins, W., & Tandon, L. (2011). Interactive image quantification tools in nuclear material forensics. *Image Processing: Machine Vision Applications Iv, 7877*, 787708. doi:10.1117/12.877319

RSD-1. (n.d.). Retrieved November 22, 2013, from Wikipedia, http://en.wikipedia.org/wiki/RDS-1

Salter, L. & Harley, J. (1965). Trinitite - cobalt-60 cesium-137 and europium-152. *Science, 148*(3672), 954-&. doi:10.1126/science.148.3672.954

Sandpaper. (n.d.). Retrieved April 13, 2014, from Sandpaper (coated abrasives) website, http://www.sizes.com/tools/sandpaper.htm

Semipalantinsk Nuclear Test Site. (n.d.). In *Images for Semipalantinsk*. Retrieved from https://www.google.com/search?q=semipalatinsk&tbm=isch&tbo=u&source=univ&sa=X&ei=7GlJU4vKFsqc8QGOxoGICw&sqi=2&ved=0CEIQsAQ&biw=1366&bih=625

SiNi Probe. (n.d.). In Budget Sensors Website. Retrieved from http://www.budgetsensors.com/downloads/SiNi_Datasheet.pdf

Stanley, F. E., Stalcup, A. M., & Spitz, H. B. (2013). A brief introduction to analytical methods in nuclear forensics. *Journal of Radioanalytical and Nuclear Chemistry, 295*(2), 1385-1393. doi:10.1007/s10967-012-1927-3

Tandon, L., Kuhn, K., Martinez, P., Banar, J., & Walker, L. (2009). Establishing reactor operations from uranium targets used for the production of plutonium. *Journal of Radioanalytical and Nuclear Chemistry, 282*(2), 573; 573-579; 579.

The College of William and Mary, Main Campus. (2010, February 17). *Radiation Safety Manual.* Retrieved from https://www.wm.edu/offices/facilities/documents/safety/radiation/RadSafetyManual-VaRadMat-2-17-10.pdf

Thormann, E., Pettersson, T., & Claesson, P.M. (2009). How to measure forces with atomic force microscopy without significant influence from nonlinear optical lever sensitivity. AIP Review of Scientific Instruments, 80, 093701. doi:10.1063/1.3194048

Trinitite. (n.d.). Retrieved March 20, 2013, from Wikipedia, http://en.wikipedia.org/wiki/Trinitite

Trinity Nuclear Test. (n.d.) Retrieved November 22, 2013, from Wikipedia, http://en.wikipedia.org/wiki/Trinity_(nuclear_test)

Trinity Test Site. (n.d.). In *Images for Trinity Test Site*. Retrieved from https://www.google.com/search?q=trinity+test+site&tbm=isch&tbo=u&source=univ&sa=X&ei=4WRJU4flOofy8QG_24HoDQ&sqi=2&ved=0CDMQsAQ&biw=1366&bih=625

Tuniz, C., Zoppi, U., & Hotchkis, M. (2004). Sherlock holmes counts the atoms. *Nuclear Instruments & Methods in Physics Research.Section B, Beam Interactions with Materials and Atoms, 213*, 469; 469-475; 475.

Ulicny, W. (2009). National technical nuclear forensics: overview, qa, and expertise development. *Domestic Nuclear Detection Office*. Retrieved from The National Technical Nuclear Forensics Center

Veeco (n.d.) A practical guide to scanning probe microscopy. Retrieved from http://128.239.100.218/member/download/Administrative/Manuals/Bruker%20Manuals/Veeco%20-%20A%20Practical%20Guide%20to%20SPM_0829_05.pdf

Veeco (2000) Basic spm training course. Retrieved from http://128.239.100.218/member/download/Administrative/Manuals/Bruker%20Manuals/MANUAL,%20BASIC%20SPM%20TRAINING,%20STANDARD%20VER-B%20(004-131-000).pdf

Wallenius, M., Luetzenkirchen, K., Mayer, K., Ray, I., & de las Heras, L. A. (2007). Nuclear forensic investigations with a focus on plutonium. *Journal of Alloys and Compounds, 444-445*, 57; 57-62; 62.

Wallenius, M., Mayer, K., & Ray, I. (2006). Nuclear forensic investigations: Two case studies. *Forensic Science International, 156*(1), 55-62. doi:10.1016/j.forsciint.2004.12.029

Wallenius, M., Mayer, K., & Varga, Z. (n.d.). Nuclear forensics - a methodology to reveal the past of nuclear materials [PDF document]. Retrieved from Defense Threat Reduction Agency

Yamamoto, M., Hoshi, M., Takada, J., Sakaguchi, A., & Apsalikov, K. (2004). Distributions of pu isotopes and cs in soil from semipalatinsk nuclear test site detonations throughout southern districts. *Journal of Radioanalytical and Nuclear Chemistry, 261*(1), 19; 19-36; 36.

Yamamoto, M., Hoshi, M., Takada, J., Sekerbaev, A., & Gusev, B. (1999). Pu isotopes and137Cs in the surrounding areas of the former soviet union's semipalatinsk nuclear test site. *Journal of Radioanalytical and Nuclear Chemistry, 242*(1), 63; 63-74; 74.

Yashvant. (2013, October 3). Typical atomic force microscope (afm) setup. *Opensource Handbook of Nanoscience and Nanotechnology*. Retrieved from http://kristian.molhave.dk