U.S. NWTRB
2017 Summer Meeting
Abstracts of Poster Presentations

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Nuclear Waste Technical Review Board (NWTRB) June 2017 Meeting Poster Abstract

Title – Correlating Aging and Durability of Pre-Viking Hillfort Glasses from the Broborg Hillfort Site, Sweden to Predicted Long-Term Performance of Vitrified Waste

Authors – Carolyn I. Pearce[1], Jamie L. Weaver[1,2], Edward Vicenzi[3], Thomas Lam[3], Tamas Varga[1], Micah D. Miller[1], Bruce W. Arey[1], Michele A. Conroy[1], John S. McCloy[4], Rolf Sjoblom[5], Eva Hjarthner-Holdar[6]. Erik Ogenhall[6], Michael J. Schweiger[1], David K. Peeler[1], and Albert A. Kruger[7]

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Abstract

The goal of this project is to determine the long term durability of Broborg Hillfort glass to support disposal of low-activity waste (LAW) glass in the Hanford Integrated Disposal Facility (IDF). This includes determining the ancient glass melting technology used so that synthetic glass of the sample composition can be generated and used in short-term accelerate aging test methods. Investigating natural and archeological glass alteration is essential to understand and predict long-term performance of nuclear waste glasses. While laboratory glass alteration experiments deliver short-term glass corrosion rates, natural and archeological analogues provide insight into long-term mechanism(s) driving glass corrosion in natural environments. These mechanisms are of interest to the nuclear waste glass community, as key radionuclides (e.g., technetium, half-life = 213,000 years) must be retained for thousands of years. For this purpose, ancient glasses found in a Swedish hillfort, containing elements used in simulated Hanford nuclear waste glass formulations, have been identified. These artifacts have been altered in a natural environment since ca. 400-575 AD. Non-destructive x-ray computed tomography (XCT) was used to select areas for sectioning, with subsequent micro x-ray diffraction (μ-XRD) to find amorphous glassy areas, micro x-ray fluorescence (μ-XRF) to determine glass chemistry and electron microscopy to analyze alteration layers on the glass surface. Characterization of these alteration layers proves a unique insight into natural process that drive glass corrosion over thousands of years. Data on two silicate glasses, one high iron and one low iron, found at the Broborg Hillfort will be presented, and the role of biodeterioration in their corrosion will be discussed.

Title – Reconstructing Vitrified Wallbuilding Technology for Broborg Hillfort Site, Sweden

Authors –John S. McCloy,[1,4] Mostafa Ahmadzadeh,[1] José Marcial,[1] Mahmood Abusamha,[1] Joseph Osborn,[1] Owen Neill,[2] Alex Iveson,[2] Joshua Feinberg,[3] Carolyn I. Pearce,[4] Jamie L. Weaver,[5] Rolf Sjöblom,[6] Eva Hjarthner-Holdar,[7] Erik Ogenhall,[7] Michael J. Schweiger,[4] David K. Peeler,[4] and Albert A. Kruger[8]

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Abstract

One primary goal of this international collaboration is to utilize the natural experiment of long term glass-ceramic degradation at the Broborg Hillfort in Sweden to inform models incorporating compositional dependence of glass corrosion. In tandem with this goal, we are supporting the work to preserve the cultural heritage and rediscover the technology of the pre-Viking group whose practices lead to the unusual natural experiments. To this end, we have investigated the micro-scale interfaces between different glass and crystalline phases in some samples from Broborg using X-ray diffraction and Electron Probe Microanalysis. Microstructures observed indicate high undercooling resulting in dendritic textures of pyroxenes, feldspars, and cordierite. Relict and newly formed spinels containing iron have also been considered as recorders of the Earth's magnetic field, and preliminary experiments using a Vibrating Sample Magnetometer have determined that glassy hillfort specimens are suitable for paleomagnetic measurements, including irrefutable tests of whether melting was due to lightning or anthropogenic heating. Experiments conducted to replicate the glassy phase, for the purposes of conducting corrosion experiments, indicated that excessively high temperatures >1450°C were required to melt compositions measured in hillfort samples as both "dark" (mafic) and "light" (felsic) glasses. These temperatures would not be achievable in antiquity, so further investigations have begun to assess the role of water content and iron redox in melt viscosity. Additionally, samples of the host rock amphibolite, thought to be the source of the mafic melt, have been obtained from Sweden and characterized for chemistry and phase. Initial investigations suggest than not all local amphibolites would have been suitable, with some having excessively high quartz content and inadequate amphibole minerals containing bound hydroxides to lower melting temperatures. Characterization of hillfort samples, simulant glasses, and amphibolite rocks will be presented. Production of simulant glasses with the right character and melting point is critical for accelerated lab corrosion testing to compare with the natural analogue glasses, exposed for ~1500 years.

Structural and leaching studies on new glass matrices developed for HLW conditioning

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One very good way of storing high-level radioactive waste produced by reprocessing spent fuel of civil nuclear reactors is to incorporate it into an inert host material. Borosilicate based glasses are generally accepted as proper HLW isolating media [1], as they are satisfying the following major requirements: the radioactive elements become immobilized as part of the host material structure; the leaching rate of radioactive elements is acceptably low, and the encapsulation cost is acceptable. Structural characterization of these glasses is essential for understanding the glass durability. Due to the high number of contributing elements and the overlapping atomic distances, however, it is very difficult to derive adequate structural data from diffraction experiments alone.

We investigated the multi-component sodium borosilicate waste glasses with the main (matrix) composition of SiO_2-B_2O_3-Na_2O-BaO-ZrO_2. Our study carried out on matrix glasses doped respectively with UO_3 and CeO_2, Nd_2O_3, the latter two used as chemical models of (Pu, Cm/Am) actinides, with the aim to clarify the correlation between structural characteristics and thermal and glass stability.

The investigated samples were prepared by melt-quenched technique [2,3]. For the study of the short- and intermediate range order we performed NMR spectroscopy and neutron- and high energy X-ray diffraction measurements. The experimental data were processed by a combination of direct sine-Fourier transformation of the structure factor, S(Q) and reverse Monte Carlo (RMC) modeling [4].

RMC simulation of the experimental S(Q) was successfully applied to generate a reliable 3-dimensional atomic configuration. Several partial atomic pair correlation functions and most of the corresponding coordination number distributions have been determined. It was established that the basic network structure consists $^{[4]}$Si-O-$^{[3]}$B, $^{[4]}$Si-O-$^{[4]}$B bond-linkages and the stable matrix can incorporate a maximum of 40 w% HLW. The RMC simulation of the neutron diffraction data is consistent with a model where the uranium ions are incorporated into interstitial voids in the essentially unmodified network structure of the starting host glass. From significant second nearest neighbor atomic pair correlations have been revealed that Ce and Nd accommodates in both silicate and borate sites.

Leachability studies were performed using various time and temperature conditions. The studied glasses possess good glass and hydrolytic stability. Details of the structural characteristics and leaching tests will be presented.

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Understanding the disposability of nuclear waste glass: Insights from the United Kingdom nuclear research programme

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The post closure safety case for the deep geological disposal of UK nuclear waste is progressing to take into account new wasteforms arising from decommissioning of nuclear sites, such as Sellafield. Of utmost importance in the safety case is an understanding of the long term performance of nuclear wasteforms, therefore, an understanding of the mechanisms and kinetics of dissolution in groundwater are required. The University of Sheffield, through the UK Engineering and Physical Science Research Council (EPSRC) funded programmes, Diamond and Distinctive, is leading the UK effort in development of novel wasteform materials for problematic decommissioning wastes and also in evaluating the dissolution of UK vitrified high- and intermediate-level waste.

This poster will give an international perspective on vitrified nuclear waste research, reviewing recent efforts to develop vitrified wasteforms for plutonium contaminated materials and decommissioning wastes, and the production of glass-ceramics for the immobilisation of plutonium residues. It will discuss recent advances made in understanding the durability of these wasteforms, particularly in the presence of high-pH, Ca-rich solutions expected under the proposed conditions of final disposal in a UK geological disposal facility, and also in determining the kinetics of dissolution, for example, through imaging techniques.

Evaluation of novel leaching assessment for nuclear waste glasses

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At the Hanford site, USA, low activity tank wastes will be immobilised by vitrification to create 150-350,000m^3 of immobilised low activity waste (ILAW). ILAW is destined for disposal in a shallow subsurface Integrated Disposal Facility (IDF) where conditions are expected to differ from those within a deep geological disposal facility for high level waste.

Current disposability assessment for ILAW glass relies upon highly accelerated leaching tests, which are not representative of the temperature or environmental conditions of the IDF. These methods are also poorly reproducible; variability between tests, even with the same operator, gives a correlation coefficient of < 0.7, which dramatically constrains the acceptable glass formulation. Project GLAD (Glass Leaching Assessment for Disposability), funded by the Department of Energy Office of River Protection Waste Treatment & Immobilization Plant Project through the EM International Program, investigates a new leaching approach for assessing the durability of ILAW glasses at relevant temperatures and conditions of disposal, using two established U.S. Environmental Protection Agency (EPA) leaching protocols. We critically appraise the application of these methodologies, through investigation of the dissolution of three ILAW glasses (LAW-A44, ORP-LB2 and LAW-A23) as a function of temperature, pH, leaching duration and groundwater composition. Results are compared with those from established leaching methodologies currently accepted in Europe for evaluation of high level waste glasses, including PCT and MCC-1 protocols. The advantages and limitations of the EPA methods are discussed and their reproducibility is assessed by comparing results across multiple laboratories.

Funding for this work was provided by William F. Hamel, Jr., Assistant Manager, of the U.S. Department of Energy Office of River Protection Waste Treatment & Immobilization Plant Project through the EM International Program.

Project collaborators: University of Sheffield, Pacific Northwest National Laboratory (PNNL) and the Consortium for Risk Evaluation with Stakeholder Participation (CRESP).

Hanford Site Focused Cementitious Waste Form Testing at PNNL

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ABSTRACT

Immobilization of Hanford Site tank wastes will occur at the Hanford Tank Waste Treatment and Immobilization Plant (WTP), which is currently under construction. The low activity waste (LAW) fraction of the waste inventory will be disposed of at the Hanford Site Integrated Disposal Facility (IDF). Secondary waste streams will be generated during vitrification within the HLW and LAW vitrification facilities and from activities such as liquid waste treatment and evaporation. The baseline technology for immobilization of the secondary waste streams, both solid and liquid forms, is low temperature cementitious waste forms (CWFs). A CWF development program focused towards immobilization of projected secondary waste streams from WTP and the Hanford Site's Effluent Treatment Facility (ETF) and Effluent Management Facility (EMF) is under way to support maintenance of the IDF performance assessment (PA) efforts. A CWF is also a potential technology for supplemental immobilization of pretreated Hanford LAW. The IDF PA provides an analysis of the long-term performance of the IDF as a basis to set requirements for CWF and facility design. Routine maintenance is an important component of PA efforts as new information and technological developments on elements of the disposal system becomes known. In this poster, we present an overview of the CWF testing program, discuss technological developments for improved CWF performance, and discuss a strategy for the CWF testing program to reduce risks and uncertainties associated with the CWF release model within the PA.

Formulation Development for Treatment of EMF Evaporator Bottoms

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ABSTRACT

The Hanford direct feed low activity waste (DFLAW) process will treat tanks supernate to remove cesium (Cs) by ion exchange followed by vitrification of the decontaminated Low Activity Waste (LAW) stream in the WTP LAW melter. The LAW vitrification facility uses two types of scrubbers to remove the contaminants from the melter off-gas waste stream. These scrubbers, the Submerged Bed Scrubber (SBS) and Wet Electrostatic Precipitator (WESP) generate an off-gas condensate aqueous stream. This melter off-gas waste stream will be evaporated in the WTP Effluent Management Facility (EMF) and the evaporator concentrates waste stream will be recycled and blended in patches with the LAW melter feed. The EMF evaporator concentrates waste steam primary radioactive contaminant is expected to be ^{99}Tc, which is present in the tank supernate and is volatile under melter high temperature conditions and the primary non-radioactive contaminants are ammonium, chloride, and fluoride ions. Recycling of this stream into the melter increases fluctuation in the waste feed composition, lowering the glass waste loading and ultimately to the production of more LAW glass.
To this end, identification and development of technologies to immobilize or remove the contaminants from this off-gas condensate stream represents an opportunity that would facilitate alternate treatment schemes and disposition options.

An EMF evaporator feed core simulant was developed, a laboratory scale evaporation of the simulant was conducted, immobilization/solidification of the EMF bottoms waste stream using multiple dry material compositions was completed and a waste form was developed. An extended screening tests to determine whether the full range of predicted EMF liquid waste concentrate compositions are a factor in producing acceptable solidified waste forms. Cementitious waste forms were produced and testing performed for a series of eight variations of the projected EMF evaporator bottoms simulants. Simulants contained varying concentrations of boron, chloride, nitrite, and sulfate, and three dry ingredient recipes containing two to three of the following ingredients: Blast Furnace Slag (BFS), Ordinary Portland Cement (OPC), Fly Ash (FA), and Aquaset II-GH (Aquaset). Simulants spiked with Zn, Cr, As, Se, Tc, I and Hg were used to prepare different formulations of cementitious waste forms. These cementitious waste forms were then subject to a series of analytical experimental tests to assess waste forms compatibility with disposal site WAC.

A Strategy for Maintenance of the Long-Term Performance Assessment of Immobilized Low-Activity Waste Glass

Joseph V. Ryan (PNNL), James J. Neeway (PNNL) Gary L. Smith (PNNL), Elvie E. Brown (WRPS), Steven E. Kelly (WRPS), and David J. Swanberg (WRPS)

ABSTRACT

The low-activity waste (LAW) fraction of waste from the Hanford site will be immobilized by vitrification at the Hanford Tank Waste Treatment and Immobilization Plant (WTP) and disposed at the nearby Integrated Disposal Facility (IDF). The Performance Assessment (PA) for the IDF provides an analysis of the long-term performance of the planned disposal system as a basis to set requirements for the waste form and the facility design. Routine updates are an important provision of the PA as new information on elements of the disposal system becomes known. In this poster, we present a strategy for testing programs to reduce risks and uncertainties within the immobilized low-activity waste (ILAW) glass corrosion model. We have identified three areas to target the most impactful remaining uncertainties in PA release rates due to glass: 1) glass composition, 2) the ion exchange reaction, and 3) secondary phase formation.

The impact from glass composition is a matter of ensuring that parameters used represent the suite of glass compositions expected to be produced. Ion exchange is currently conservatively represented by a constant release of sodium, whereas a diffusive term would be more technically appropriate. The impact of secondary phases range from insignificant to impactful, so the use of a representative suite of phases in geochemical modeling is important. The reduction of uncertainties in the PA will decrease conservatism in existing assumptions while increasing the overall technical basis of each parameter.

Current efforts in immobilized low-activity waste glass testing to support the Hanford site Integrated Disposal Facility

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ABSTRACT

Immobilized low-activity waste (ILAW) glass produced at the Hanford Tank Waste Treatment and Immobilization Plant (WTP) will be disposed of at the Hanford site Integrated Disposal Facility (IDF). In order to inform models used to predict the performance of ILAW glass at IDF, it is necessary to understand the dissolution mechanism and corrosion behavior of the glass in a variety of conditions. In this poster, we discuss a variety of tests that are used to inform the models that account for the effects of 1) solution chemistry, 2) the ion exchange reaction, and 3) secondary phase formation on the glass dissolution rate. To quantify the effects of solution chemistry, specifically changes in pH, T, and Si concentration, the single-pass flow-through (SPFT) test method is applied. The results from this test are used to parameterize a kinetic rate model of glass dissolution that is based on transition-state theory. Furthermore, SPFT tests that are conducted with relatively high concentrations of Si are used to determine an ion exchange rate, an important mechanism that is theorized to control the long-term dissolution rate of glass. Additionally, tests such as the pressurized unsaturated flow (PUF) test method, long-term product consistency tests (PCT), and field lysimeters are capable of providing a suite of secondary phases that are likely to form as the glass corrodes. The collection of kinetic rate law parameters, the ion exchange term, and the suite of secondary phases are provided to IDF performance assessment (PA) modelers to underpin the technical credibility of the PA model.

Field Scale Lysimeter Studies to Test Glass and Cementitious Waste Form Performance

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Field-scale experiments are used to validate the models used in the near-field simulation of the Hanford Site Integrated Disposal Facility (IDF) Performance Assessment (PA). The goal of this scope is to use the Hanford site field lysimeter test facility (FLTF) to replicate the long-term field conditions anticipated at the IDF. This will be accomplished by simulating important aspects of waste form degradation and the interaction of waste form corrosion products with the surrounding IDF disposal site materials and soils. The lysimeter studies will examine cementitious and glass waste forms targeted for disposal within the Hanford Site IDF, as well as potential impacts from co-disposal. An integrated laboratory-field-modeling approach will be used to assure that the experiments yield data that validate important aspects of waste form release and subsequent transport of contaminants through the unsaturated zone. Lysimeter field scale studies will utilize the state of the art FLTF facility to validate existing models used in near-field simulation of the IDF PA, generate a set of parameters needed for modeling long-term performance of waste forms, and significantly reduce uncertainty in PA predictions.

Dissolution of Borosilicate Glass into Brine Solutions: Effects of Sodium and Magnesium

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Flow-through experiments were carried out at 90 °C and pH(25 °C) = 9 determine the dissolution rates of the International Simple Glass (ISG—a six oxide borosilicate glass) and to elucidate the effects of Na^+ and Mg^{2+} on the reaction kinetics. Understanding the impact of these two cations in solutions is important because they are the most common cations in brine solutions associated with rock salt and in brines resulting from repeated cycles of evaporation-precipitation near heat-producing waste. In addition, experiments conducted systematically over NaCl or $MgCl_2$ concentration intervals enable us to test the hypothesis that borosilicate glass dissolution rates are governed by two competing mechanisms: At low NaCl concentrations rates are catalyzed by Na^+, much in the same way that SiO_2 polymorphs react in electrolyte solutions, and that in more concentrated NaCl solutions rates decrease in response to diminishing activities of water, according to the following rate expression:

$$Rate = k_0 \cdot (a_{SiO_2}) \cdot (a_{H_2O})^2 \cdot \left(1 - \frac{Q}{K}\right) \quad \text{[Eq. 1]}$$

where k_0 is the rate constant, a is the activity of the subscripted species, Q is the ion activity product and K is the solubility constant for the rate-determining solid. Powdered samples and monoliths (15 × 15 × 4 mm coupons) were run at identical q/S (flow-rate to surface area ratio) conditions. Experiments were run in duplicate over NaCl (0 to 4.0 m NaCl) and $MgCl_2$ (0 to 1.5 m $MgCl_2$) concentration intervals. An additional set of ISG dissolution experiments were run in a simple mixed brine solution (GWB; 3.5 m NaCl, 1.0 m $MgCl_2$). Dissolution rates were determined by steady-state release of Si and B to effluent solutions and by measuring the surface retreat rate using the interferometric method on monoliths. Dissolution rates based on Si release in NaCl- and $MgCl_2$-free solutions vary between ~0.2 and 0.7 g/(m^2·d). In NaCl solutions the rate of dissolution increases by a factor of ~10× upon addition of low concentrations (0.03 molal) of NaCl. Dissolution remains constant at these elevated rates in NaCl solutions up to ~1.0 molal NaCl, and then decrease. The rate decrease is consistent with that expected from Eq. 1: The rate decreases as a function of the activity of water squared. In $MgCl_2$ solutions, the presence of Mg^{2+} has the opposite effect: Rates decrease (by a factor of ~10×) with the addition of $MgCl_2$, even at low concentrations. Rates determined by surface retreat are roughly equal to those determined by chemical assay of effluent solution. Collectively, these results point toward a clear description of borosilicate glass dissolution based on simple TST-like constructs and hint that the principal mechanism governing dissolution is rupture of the Si—O bond.

ALTGLASS Database: An Informatics Tool for Glass Durability Modeling

Cory L. Trivelpiece, Carol M. Jantzen, Charles L. Crawford

The Accelerated Leach Testing of GLASS (ALTGLASS) database is a compilation of data collected from decades of high-level (HLW) and low-activity (LAW) nuclear waste glass accelerated durability testing. The database contains both short and long term product consistency test (PCT-A and B) results covering chemical analyses of the corroding solutions as well as solids analysis of the remnant glasses and secondary phases that may have resulted from corrosion processes when available. The most current version, ALTGLASS v. 3.0, contains entries for over 525 glasses. Input data for ALTGLASS has been generated at Savannah River National Laboratory (SRNL) but also includes submissions from other U.S. national laboratories, university entities, and international collaborative partners, such as the National Nuclear Laboratory in the United Kingdom. Researchers at SRNL originally developed and currently maintain custodianship of the database, which is available upon request in electronic format to both national and international research groups working on the long-term durability of nuclear waste glass. This poster will demonstrate the utility of ALTGLASS by highlighting key facts about the database in addition to recently published applications of the database, which could lead to a paradigm shift in the approach to geologic repository model development.

Center for Performance and Design of Nuclear Waste Forms and Containers (WastePD) an Energy Frontiers Research Center

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The US Department of Energy recently awarded funds to create the Center for Performance and Design of Nuclear Waste Forms and Containers (WastePD) as part of the Energy Frontier Research Center (EFRC) program. WastePD is a focused scientific effort to understand the details of and commonalities in the aqueous corrosion mechanisms of glass, ceramics, and metals. For each material class, the corrosion mechanism involves the formation of a thin surface film through atomic scale interactions with the environment. Synergistic interactions between researchers in these areas will result in scientific advances for these very complicated problems that would not otherwise be possible. Significant progress in the area of glass corrosion understanding has been developed in the first six months of the WastePD Center.

The *WastePD Glass Team* is developing a fundamental understanding of the structure and chemistry of the reacting glass-solution interface and determining the dominant mechanism of glass corrosion and the impact of environmental and chemical parameters. Initial work has focused on combined testing and modeling of a six-component glass, International Simple Glass (ISG), selected by the international community to build a consensus on the understanding of glass corrosion processes. Using MD simulations, both the bulk and surface structures of the ISG were analyzed in detail. The atomic structure of ISG was generated and structure-related properties such as pair distribution function, coordination number, bond angle distribution, etc. were calculated. Zr^{4+} was found to be mainly six-fold coordinated and $[ZrO_6]^{2-}$ units in the glass are preferentially charge-compensated by Ca^{2+} ions. B^{3+} has a mixed three- and four-fold coordination and was found to play an important role in connecting the Si/Al network, which has profound implications for glass corrosion and gel layer formation. Using reactive force field MD, the interaction between nano-porous structures simulating passivation layers and water was studied. Several structures with cylindrical pores were generated to give insights into fundamental mechanisms controlling water reactivity at the atomic scale and to help interpret experimental results. The properties of passivating layers formed on the ISG were studied by TGA, NMR and ToF-SIMS in the absence and presence of chloride salts. Water content and speciation of nano-confined water were determined, and the mobility of the water molecules within the passivating layer was assessed for the first time using isotopically tagged water molecules and ToF-SIMS depth profiling. A stoichiometry-based structural model for oxygen speciation on the ISG surface was developed using XPS characterization of the surfaces of glass samples and the coordination number of each network-former element predicted from MD simulations. Specular reflection IR was used to analyze the silicate network structure of the altered surface layer of reacted ISG and control surfaces. Samples treated in LiCl solution showed dramatic differences compared to those exposed in a KCl solution. The techniques needed for flash-freezing and cryogenic FIB sectioning were developed and applied to corroded glass. After solving significant experimental complications, the first-ever APT characterization of a cryogenically prepared, site-specific, lift-out specimen was successfully achieved. This method allows for characterization of the altered surface layer and environment without artifacts from sample preparation.