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Evaluation of Uranium-235 Measurement Techniques

May 2017

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MW Dibert

U.S. DEPARTMENT OF
ENERGY

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Richland, Washington 99352

Summary

Alternative fuel compositions and forms are being explored to meet the U.S.[1] and European[2] goals to convert civilian research nuclear reactors from highly enriched uranium (HEU, ^{235}U typically > 90 wt%) to low-enriched uranium (LEU, ^{235}U < 20 wt%) fuel. For high-power applications such as U.S. high-performance research reactors, metallic LEU fuels with higher ^{235}U density than that of U_3Si_2 must be developed. Extensive studies of metallic γ-U-Mo alloys as dispersed particles embedded in Al 6061 have found that at higher operating temperature and higher burnup, interfacial reactions occur between the U-Mo particles and the Al alloy matrix to form uranium aluminides. This leads to undesirable swelling or "pillowing" of the dispersion fuel plates. As an alternative design that reduces the contact area between U-Mo and Al, and simultaneously increases the ^{235}U density, monolithic γ-U-Mo fuel plates are being developed that employ a Zr barrier between the Al and U-Mo.[3] Monolithic U-Mo fuel plates are rolled to final fuel element form from the original cast ingot, and thus any inhomogeneities in ^{235}U distribution present in the cast ingot are maintained, and potentially exaggerated, in the final fuel foil. The tolerance for inhomogeneities in the ^{235}U concentration in the final fuel element foil is very low. A near-real-time, nondestructive technique to evaluate the ^{235}U distribution in the cast ingot is required in order to provide feedback to the casting process. No technique that provides fine spatial resolution and accurate quantification of ^{235}U concentration is currently available as a commercial off-the-shelf system.

The methods to measure isotopic abundance rely on one of two basic principles to distinguish the isotopes of uranium: mass spectrometry or spectroscopy of radioactive emission signatures. Inductively coupled plasma–mass spectrometry (ICP-MS) and thermal ionization mass spectrometry both utilize mass spectrometry to analyze ionized species in the gas phase. Both are destructive techniques and require significant sample preparation before measurement; thermal ionization mass spectrometry has the most onerous sample requirements but is also considered the most accurate measure of ^{235}U content. An alternative analysis method using mass spectrometry is laser ablation-ICP-MS (LA-ICP-MS). LA-ICP-MS requires no sample preparation, provides high spatial resolution, and is only minimally destructive. In contrast, methods to measure radioactive emission signatures are fully nondestructive. The most well-established technique is gamma spectroscopy. Using the enrichment meter method, gamma spectroscopy can achieve high accuracy when measuring ^{235}U content. Non-spectroscopic techniques such as autoradiography can be performed very quickly, and inherently provide a spatially resolved map of radiation intensity, but significant development would be required to utilize autoradiography to distinguish between ^{235}U and ^{238}U. Active neutron detection techniques, in which a neutron source is placed behind the sample of interest, are not well developed for uranium, and further, would offer little or no spatial resolution.

Based on the technical analysis herein, gamma spectroscopy has been recommended to provide a near-real-time measure of the ^{235}U distribution in U-Mo cast plates. Sufficient spatial resolution can be obtained with a small aperture and collimator, which allows measurement of a small area. From measurements of multiple areas, a map of ^{235}U distribution can be made. ICP-MS and LA-ICP-MS will be

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[1] Keiser DD, SL Hayes, MK Meyer, and CR Clark. 2003. "High-density, low-enriched uranium fuel for nuclear research reactors." JOM-J Miner. Met Mater Soc. 55(9):55–58.

[2] Van den Berghe S and P Lemoine. 2014. "Review of 15 years of high-density low-enriched UMo dispersion fuel development for research reactors in Europe." *Nucl. Eng. Technol.* 46(2):125–146.

used as needed for benchmarking. Thermal ionization mass spectrometry may also be employed to characterize the ^{235}U standards that will be fabricated to support the gamma spectroscopy measurements.

Acronyms and Abbreviations

COTS	commercial off-the-shelf
FOV	field of view
HEU	highly enriched uranium
HPGe	high purity germanium
ICP	inductively coupled plasma
ICP-MS	inductively coupled plasma mass spectrometry
LA-ICP-MS	laser ablation inductively coupled plasma mass spectrometry
LA-MC-ICP-MS	laser ablation multi-collector inductively coupled plasma mass spectrometry
LANL	Los Alamos National Laboratory
LEU	low-enriched uranium
MC-ICP-MS	multi-collector inductively coupled plasma mass spectrometry
MGA	multigroup analysis
MS	mass spectrometer
NaI	sodium iodide
NIST	National Institute of Standards and Technology
PNNL	Pacific Northwest National Laboratory
TIMS	thermal ionization mass spectrometry
Tl:NaI	thallium-doped sodium iodide
U_3Si_2	uranium silicide
USHPRR	U.S. high-performance research reactors
Y-12	Y-12 National Security Complex, Oak Ridge, TN

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1.0 Introduction

Alternative fuel compositions and forms are being explored to meet the U.S. (Keiser et al. 2003) and European (Van den Berghe et al. 2014) goals to convert civilian research nuclear reactors from highly enriched uranium (HEU, ^{235}U typically > 90 wt%) to low-enriched uranium (LEU, ^{235}U < 20 wt%) fuel. Uranium silicide (U_3Si_2) fabricated from LEU was qualified in the 1990s, and most lower-power research reactors have been converted to this fuel. However, as uranium enrichment is decreased, the net fissile (^{235}U) atom density also decreases, and U_3Si_2 does not possess sufficient ^{235}U density for use in U.S. high-power research reactors (USHPRR). For high-power applications such as USHPRR, metallic LEU fuels with higher ^{235}U density must be developed.

Metallic uranium exhibits higher radiation tolerance in the high temperature (>775°C) γ-U phase with the body-centered cubic crystal structure (Perez et al. 2016), which is more malleable and ductile than the low temperature orthorhombic-structure α-U phase. Alloying with 6–10% Mo stabilizes the γ-U phase as the material is quenched from high temperature. Extensive studies of γ-U-Mo alloys as dispersed particles embedded in Al 6061 have been undertaken (Keiser et al. 2003, Van den Berghe et al. 2014, Champion et al. 2014, Kim et al. 2016, Leenaers et al. 2016). It was found that at higher operating temperature and higher burnup, interfacial reactions occur between the U-Mo particles and the Al alloy matrix to form uranium aluminides. This leads to undesirable swelling or “pillowing” of the dispersion fuel plates. As an alternative design that reduces the contact area between U-Mo and Al, and simultaneously increases the ^{235}U density, monolithic γ-U-Mo fuel plates are being developed, employing a Zr barrier between the Al and U-Mo (Clarke et al. 2015).

In dispersion fuels, the cast U-Mo ingot is ground or atomized to generate a fine powder, which is mixed and dispersed in the Al matrix. This processing homogenizes any nonuniformity in ^{235}U distribution present in the original casting. In contrast, monolithic U-Mo fuel plates are rolled to final fuel element form from the original cast ingot, and thus any inhomogeneities in ^{235}U distribution present in the cast ingot are maintained, and potentially exaggerated, in the final fuel foil. The tolerance for inhomogeneities in the ^{235}U concentration in the final fuel element foil is very low: the operators of the High Flux Isotope Reactor at Oak Ridge National Laboratory estimate that the ^{235}U concentration (as a fraction of total U content) must not vary more than ±0.1 wt% in a 1×1 mm^2 area of 0.012” (300 μm) foil.

The LEU U-Mo ingots cast at the Y-12 National Security Complex are fabricated from the appropriate nominal amounts of HEU (93 wt% ^{235}U) and depleted uranium-Mo alloy (12.5 wt% Mo) to achieve a target LEU U-Mo composition of 19.75 ± 0.2 wt% ^{235}U and 10 wt% Mo. Note that the target enrichment value of 19.75% ^{235}U is very close to the regulatory cutoff of 20%; at concentrations ≥ 20% ^{235}U, the material must be treated as HEU. Furthermore, the thermodynamic driving force to mix ^{235}U and ^{238}U in the casting process is very low due to their chemical similarity, and significant inhomogeneities in ^{235}U distribution have been observed in the final U-Mo cast fuel plates. These as-cast plates measure 8" × 9" × 0.2" thick, and will be subjected to a series of hot-rolling and cold-rolling steps to generate the final fuel foils measuring 3" × 48" × 0.012" (300 μm) thick.

To confirm both the enrichment and the homogeneous distribution of ^{235}U in the cast plates, it is necessary to measure the spatial distribution of ^{235}U. An established, industry-standard method for this evaluation does not exist. Therefore, we have undertaken an assessment of existing techniques that have the potential to measure the ^{235}U concentration in a spatially resolved manner.

2.0 Technology Requirements and Candidates

Ideally, the technology chosen to provide a measure of ^{235}U concentration and distribution will be nondestructive and non-intrusive, accurate, simple to use, and compatible with installation in the analytical laboratory of a manufacturing facility such as Y-12. This will allow an evaluation of the as-cast U-Mo plates in near-real-time, without the time and expense of shipping to an offsite location. Compatibility in this case requires a technique that can be operated and maintained by nonexpert personnel. Commercial off-the-shelf (COTS) technology is preferred, requiring little or no development.

The required measurement accuracy places a significant restriction on the choice of technology. The measurement technique must precisely evaluate the ^{235}U concentration to within ±0.1–0.2 wt% on a spatial scale of millimeters. This is a challenging requirement for simple, inexpensive measurement technologies. To identify the technology that offers the best combination of simplicity and accuracy, we have evaluated several candidate techniques. Although these techniques vary widely in the method of implementation, they all rely on one of two basic principles to distinguish the isotopes of uranium (Smith 1991):

- mass spectrometry
- spectroscopy of radioactive emission signatures.

Mass spectrometry is a well-established method to separate ionized gaseous atoms and molecules based on their mass. The signal collected as a function of mass is proportional to the amount of ionized species of a given mass that is present. Mass spectrometers have sufficient sensitivity to easily detect the mass difference between different isotopes of the same element, such as ^{235}U and ^{238}U. Alternatively, the radioactive signature properties of U can be exploited by measuring the gamma (high-energy photon), alpha (helium nucleus), beta (electron or positron), or neutron emission from the sample of interest.

Uranium is a gamma emitter with distinct spectroscopic signatures for each U isotope, and various spectroscopic and total-count gamma radiation detection techniques are well established. The passive alpha particle emission energies for ^{238}U and ^{235}U are similar (4.15 MeV vs. 4.3 MeV), and thus current alpha-detection techniques such as alpha cameras (Miller et al. 2014, 2015) cannot distinguish the isotopes. The neutron emission rate from ^{238}U is five orders of magnitude lower than that of ^{240}Pu (Rackham et al. 2008), which makes passive neutron detection techniques such as coincident neutron emission challenging (Rackham et al. 2008). Active neutron techniques, in which a neutron source is placed behind the sample of interest, show some potential to distinguish uranium isotopes (Ensslin et al. 2007) Mass spectrometry-based techniques are discussed in Section 2.1; Gamma spectroscopy techniques are presented in Section 2.2, non-spectroscopic gamma detection methods are discussed in Section 2.3, and neutron-based measurements are reviewed in Section 2.4.

2.1 Mass Spectrometry-Based Techniques

2.1.1 Thermal Ionization Mass Spectrometry

Thermal ionization mass spectrometry (TIMS) is considered the most accurate and precise measurement method to determine isotopic composition (Mathew et al. 2015). A small quantity (nanograms) of the material of interest is dispersed on a small ribbon filament. The filament is then resistively heated in a vacuum environment to evaporate the sample material. A small fraction of the material will evaporate as ions, not atoms; these ions are directed with electrostatic lenses to a mass spectrometry (MS) system. TIMS is a destructive measurement and requires intensive sample preparation to separate the element of interest for isotopic analysis from the matrix; it must be introduced onto the ribbon filament in an elementally pure form (Noyes et al. 2016). Care must be taken during this separation (often accomplished with some form of gas or liquid chromatography) that isotopic fractionation does not occur (i.e., the isotopic composition of the purified material must be the same as the composition of the original sample). Because TIMS requires extensive sample preparation and long measurement times, it is considered a low-throughput technique.

2.1.2 Inductively Coupled Plasma Mass Spectrometry

Inductively coupled plasma mass spectrometry (ICP-MS) is a well-known variant of ICP-based techniques to precisely measure atomic concentrations. Unlike ICP coupled with optical emission spectroscopy (ICP-OES, sometimes referred to as atomic emission spectroscopy (ICP-AES)), ICP-MS is sensitive to elemental isotopes. In ICP-MS, the inductively coupled plasma is generated in flowing argon in the center of a series of concentric quartz tubes known as the plasma torch. An initial electric spark ionizes a small portion of the argon atoms, and the charged species generated (Ar^+ and free electrons) are rapidly oscillated in the radio frequency field produced by a copper induction coil wrapped around the tubes. As the charged species oscillate, they collide with neutral Ar atoms, generating more charged species and resulting in a self-sustaining plasma. In this hot plasma (temperature of 6000–10,000 K), atoms introduced from the sample of interest are efficiently ionized. As the positive ions generated in the plasma flow out of the torch, they are directed to a mass spectrometer (MS). In a quadrupole MS, radio frequency voltages are applied to the opposing pairs of rods in the quadrupole, and for a given voltage ratio, only one mass (or, more accurately, one mass/charge ratio) can pass through the quadrupole to the detector. As the voltage ratio is swept, the detector collects a mass-dependent signal. A magnetic mass filter is required to produce precise isotopic measurements (Albarede et al. 2004). Higher resolution is achieved with an electrostatic analyzer in conjunction with a magnetic sector MS, in which the magnetic sector is a bend magnet that separates charged ions on the basis of their mass/charge ratio. With a magnetic sector MS, multiple Faraday cup collectors are employed to detect multiple mass beams simultaneously, increasing precision. These systems are referred to as multi-collector ICP-MS (MC-ICP-MS).

To prepare solid samples such as U-Mo for ICP-MS, a small portion of the sample is fully dissolved in acid (such as HNO_3), then introduced into the plasma torch via a nebulizer, or alternatively the solid sample is directly vaporized using electrothermal vaporization at the entrance to the plasma torch. Either way, the solid sample vaporizes and enters the plasma region as a gas. The technique does not possess any inherent spatial resolution; the entire sample that is introduced to the plasma torch is measured at once. Obtaining material for the measurement is destructive to the sample. However, only a very small amount of material is required, which opens the possibility of measuring material from multiple locations on the same sample to obtain spatial distribution information.

For an accurate isotope ratio measurement, it is recommended (Yang 2009) that the element of interest be separated from the matrix. This is especially important for liquid-phase samples. A drawback to ICP-MS is that impurities can be introduced during sample preparation and measurement, which can affect the accuracy of the results. Obviously, the Ar content of the sample cannot be measured since the carrier gas in the plasma is Ar. In a material system such as U-Mo, these impurity effects are not expected to have a significant effect on the measurement results. However, the accuracy of isotope ratio determination by ICP-MS is limited primarily by mass bias effects, which will affect the $^{235}U/^{238}U$ measurement. The origins of mass bias effects are not well understood but likely arise from physical phenomena occurring in the ICP-MS instrument, such as supersonic expansion of the ions and space charge effects in the zone of electric-field-induced ion acceleration. Both of these effects favor transmission of the heavier isotope into the MS, leading to nonuniform sensitivity to different isotopes of the same element. Careful mass bias correction (Yang 2009) can lead to isotope ratio measurements with precision reaching 0.001%, rivaling the precision of TIMS (Albarede et al. 2004, Yang 2009).

2.1.3 Laser Ablation ICP-MS

An alternative method to introduce solid material into the ICP-MS instrument for analysis is to utilize laser ablation. The material to be sampled is placed in an evacuated chamber in which He carrier gas is flowing. Laser ablation focuses a pulsed, high-energy laser beam with a wavelength in the ultraviolet region on the surface of the material. When the high-energy laser pulse hits the material, a complex set of absorption and energy transfer processes occurs, leading to ejection of material in a plasma or “plume.” The properties of this ablated material depend on the laser energy and pulse length (nanosecond or femtosecond). The aerosolized material is carried from the chamber within the He carrier gas, mixed with Ar gas, and introduced to the ICP digester. The ICP-MS setup can then be the same as described above, with the highest isotopic resolution obtained with a magnetic sector MS with multi-collector detection (MC-ICP-MS) (Duffin et al. 2013, 2015).

Laser ablation-ICP-MS (LA-ICP-MS) offers distinct advantages over traditional ICP-MS. The most obvious is the ease of sample preparation. With no preparation beyond surface cleaning, the material can be introduced into the ablation chamber. This also significantly reduces or eliminates the impurities that would be introduced during sample preparation for traditional ICP-MS. In addition, LA-ICP-MS offers elemental data in a spatially resolved manner. The focused laser spot is typically small (on the order of tens of square microns); rastering the laser beam can generate well-resolved maps of the elemental composition or isotopic ratio in an area of, for example, 1 mm^2 in a couple of hours. Other rastering schemes, such as line scans or sampling a series of widely spaced points are also feasible. LA-ICP-MS is a destructive technique in the sense that some material is removed during the ablation process. However, the amount of material that is removed is very small; each laser pulse creates a crater with lateral dimension approximately equal to the spot size of the laser, with a depth that is on the order of nanometers. Multi-collector systems (LA-MC-ICP-MS) offer superior sensitivity and accuracy for U isotope measurements, as discussed above. However, these detectors are tuned to the element of interest (for example, U), and cannot measure other masses such as Mo at the same time. In contrast, single-collector systems trade some isotopic resolution and accuracy for the ability to “peak-hop” between U and Mo masses, allowing data from both elements to be collected from the same ablation event.

2.2 Gamma Spectroscopy-Based Techniques

2.2.1 Gamma Spectroscopy of U

Uranium is a weakly radioactive material that primarily emits alpha particles; the half-life of ^{238}U is 4.5×10^9 years and that of ^{235}U is 7.1×10^8 years. In addition, ^{235}U emits gamma rays at 185.7 keV. ^{238}U does not directly emit gamma rays, but characteristic gamma rays are emitted by the decay chain product ^{234m}Pa at 1001.0 keV. Assuming the ^{238}U decay chain $^{238}\text{U} \rightarrow {}^{234}\text{Th} \rightarrow {}^{234m}\text{Pa}$ has reached secular equilibrium, the gamma emission of ^{234m}Pa can be used to calculate the amount of ^{238}U present.

Gamma spectroscopy is a nondestructive technique that measures the energy and intensity of gamma rays naturally emitted from radioactive materials such as U to generate a gamma spectrum. In COTS gamma spectroscopy systems, either scintillation or semiconductor detectors are employed. Scintillation detectors are typically large single crystals (i.e., 4" × 4" × 16") of sodium iodide, doped with thallium (Tl:NaI). The interaction of incident gamma rays with Tl:NaI produces electron-hole pairs in the crystal. Recombination of these electron-hole pairs produces a cascade of photons with energies that are at the blue end of the visible spectrum; the number of electron-hole pairs, and thus photons, produced is proportional to the energy of the incident gamma ray. A photocathode or photomultiplier tube converts the photons to electrical pulses, which are used to create the gamma-ray spectrum. NaI detectors are relatively inexpensive and reasonably sensitive to low gamma-ray fluxes in a short measurement time, but the resulting gamma-ray spectrum exhibits poor energy resolution as a consequence of the inherent properties of the scintillation phenomena. For this reason, quantification of ^{235}U using NaI gamma-ray spectroscopy is also of lower precision, 0.6% (ASTM 2008). The interaction of gamma rays with semiconductor detectors such as high purity germanium (HPGe) is similar to that with scintillation detectors: electron-hole pairs are generated in the detector. In an HPGe detector, these electron-hole pairs are directly collected as electrical current by applying a voltage across the detector. Again, the number of electron-hole pairs produced, and thus the measured current, is proportional to the energy of the incident gamma radiation. HPGe detectors operate at cryogenic temperatures, and offer high resolution with a precision of better than 0.2% (ASTM 2008).

Gamma spectrometers have no inherent spatial information; gamma rays are collected from the entire field of view (FOV) of the detector and an aggregate signal is produced. However, external apertures and/or collimators can be employed to significantly reduce the FOV of the detector. The FOV can be reduced to, for example, 1 mm, and data can be collected in a grid pattern with 1 mm spacing across the material of interest to generate a map. Note that, as the FOV is reduced, the measurement time must be concurrently increased to maintain sufficient counting statistics.

2.2.2 Gamma Spectroscopy Using Enrichment Meter Method

The most precise method to determine the ^{235}U content with gamma spectroscopy is termed the enrichment meter method (ASTM 2008); accuracies of better than ±0.015 wt% can be achieved (Yucel et al. 2015). In this method, the absolute peak area of the 185.7 keV gamma peak from ^{235}U is measured, and compared to well-defined reference standards to determine the amount of ^{235}U present. This method assumes that the enrichment ratio ($^{235}\text{U}/^{238}\text{U}$) is linearly proportional to the peak area (or count rate) of the ^{235}U gamma peak, and thus no information is collected from ^{238}U or its decay products. The proportionality constant to relate the ^{235}U peak to the level of enrichment is determined by measurement of reference standards that are as similar as possible to the measured material. In order for this method to achieve accurate results, both the reference standards and the material to be measured must be quasi-infinitely thick, meaning thicker than the depth at which emitted gamma rays are fully attenuated and do not escape the material to reach the detector. For uranium oxides (UO_2 and U_3O_8), this depth is estimated to be >2.7 cm (Yucel 2015), but for metallic uranium, which is more dense, the depth at which total attenuation occurs is expected to be thinner.

2.2.3 Gamma Spectroscopy, Standardless

In the absence of well-characterized reference standards for ^{235}U, alternative methods can be used to calculate the ^{235}U/^{238}U ratio from the measured gamma-ray spectrum without resorting to standards for quantification. Several different approaches to the spectral analysis have been investigated. In the most conceptually simple approach, the area ratio of the ^{235}U line at 185.7 keV to the ^{234m}Pa (decay product of ^{238}U) line at 1001.0 keV is calculated to determine the ^{235}U/^{238}U ratio. The gamma emission probabilities and half-lives of the isotopes must be included in the analysis; for ^{235}U and ^{238}U, these quantities are well established. However, the analysis is made more challenging because the two gamma peaks are far separated in energy, and so it cannot be assumed that the detector efficiency is the same for the two lines. Instead, the detector efficiency needs to be determined, either by measuring multi-element calibration standards, or by directly calculating it from the known gamma-ray emission probabilities of several ^{234}U, ^{235}U, and ^{238}U peaks in the spectrum. This calculation can be done with modeling codes such as FRAM (Sampsom 2003) and MGAU (Gunnik et al. 1990); the shape of the detector efficiency as a function of energy is not known but a functional form must be chosen by the user. Optimized methods can result in calculated enrichment values that are within 0.2–0.8 wt% (~1–4% relative) of the stated enrichment of reference standards with around 20% ^{235}U (Korob and Nuno 2006, Ruhter et al. 2001).

2.3 Gamma Detection Techniques

2.3.1 Large-Area Autoradiography

Radiography is simply the spatially resolved detection of high-energy photons (x-rays and gamma rays) or particles. Autoradiography creates an image using the alpha, beta, and gamma rays naturally emitted from a source material, in contrast to transmission radiography methods that use an external source of x-rays that pass through the material of interest (for example, medical x-ray imaging). The key difference between autoradiography and gamma-ray spectroscopy is that the radiography detector does not collect information on the energy of the incident gamma rays. Instead, each pixel of the imaging detector (typically a phosphor) undergoes physical changes (e.g., oxidation of Eu^{2+} to Eu^{3+}) that are proportional to the number of gamma rays incident on the pixel. After exposure to the material of interest for a given time, the phosphor detector is removed and "read out" using, for example, a HeNe laser (635 nm) incident on each pixel. The incident laser light reduces Eu^{3+} back to Eu^{2+} with the concurrent release of a photon. For each pixel, the number of photons generated during read-out is proportional to the number of Eu ions that were initially oxidized by incident radiation. Collection of the generated photons with a photomultiplier tube generates a gray-scale image in which brighter contrast corresponds to areas exhibiting increased levels of radiation. Exposure times must be tailored to prevent saturation of the phosphor, which occurs when all the available Eu ions in a given pixel have been oxidized. The spatial resolution of autoradiography is determined primarily by pixel size, which can be on the order of tens of microns (Deves et al. 2012). Recently, autoradiography detectors have been developed that use scintillators as the active material and a charge-coupled device camera for light detection (Miller et al. 2015).

Autoradiography inherently offers little or no chemical or isotopic specificity. The technique is quite useful for identifying the location of radioactive material, such as uranium-bearing minerals in geological samples (Deves et al. 2012, Sardini et al. 2016), but it is not well suited to analysis of ^{235}U/^{238}U distribution. Both ^{235}U and ^{238}U (via its decay products) produce gamma rays, as well as alpha particles during decay. Thus, the autoradiography image of a sample of ^{235}U looks nearly identical to that of a sample of ^{238}U (Brumbach et al. 1979). However, it may be possible to develop a shielding scheme to block the higher-energy gamma rays from ^{238}U, increasing sensitivity to ^{235}U. Quantitative analysis of the ^{235}U content at the desired precision does not appear to be feasible with autoradiography.

2.4 Neutron-Based Techniques

The neutron emission rate of uranium is too low to reasonably measure passive coincident neutron emission. As an alternative, active neutron methods such as active neutron multiplicity counting (Ensslin et al. 2007) use an external source of neutrons (typically an ^{241}Am-Li source) in an active well coincidence counter to induce fissions of ^{235}U. Coincident detection of the emitted neutrons by several neutron detectors allows the ^{235}U mass to be determined. The overall accuracy of ^{235}U enrichment was estimated to only be 4.4% relative (±0.9 wt% at 20% enrichment) (Dolan et al. 2014, Parker et al. 2016). With the low overall neutron emission rate, and particularly in the restricted geometry required for coincidence counting, it is not clear how spatially resolved information could be obtained.

3.0 Analysis of Alternatives

The technologies identified in Section 2.0 were evaluated against specific criteria identified as necessary for the characterization of LEU U-Mo cast plates and foils during processing in a manufacturing environment. These criteria include technical benchmarks such as spatial resolution and accuracy of ^{235}U enrichment measurement, and operational benchmarks such as cost and ease of use. The resulting assessment of technologies is given in Table 1, and the individual criteria are discussed in more detail below.

Table 1. Assessment of Criteria for Techniques to Measure the ^{235}U Enrichment and Spatial Distribution in U-Mo Alloys. Shading: red = does not meet criterion; yellow = possibly meets criterion; green = expected to meet criterion.

Technology	Accuracy$^{(a)}$	Spatial Resolution$^{(a)}$	Depth of Analysis	Cost$^{(b)}$	Sample Prep Required	Measurement Time$^{(c)}$	Operational Complexity$^{(d)}$	Development Required
TIMS	🟩 ±0.001 wt%	🟨 Centimeters or millimeters	Depends on sampling location	Medium	Destructive drilling, elemental separation	Long	Complex	None
ICP-MS	🟩 ±0.03 wt%	🟨 Centimeters or millimeters	Depends on sampling location	Medium	Destructive drilling, dissolution of shavings	Intermediate	Intermediate	None, Y-12 already using
Laser ablation (LA)-ICP-MS	🟩 Best: ±0.005 – 0.01 wt%;	🟩 Microns	Nanometers	High	None	Fast to intermediate, depending on # of spots	Complex	None, already done at LANL$^{(e)}$
Large-area autoradiography	🟥 Qualitative only	🟩 Microns	Millimeters	Low	None	Fast	Simple	Develop filters or detectors to provide ^{235}U sensitivity
Gamma spectroscopy, enrichment meter method	🟩 ±0.015 wt% with good standards	🟩 Expected to be millimeters	Millimeters, tomography	Low (HPGe > NaI)	None	Intermediate to long, depending on # of spots	Simple	Aperture / collimator; reference standards
Gamma spectroscopy, standardless	🟨 ± <1.0 wt%	🟩 Expected to be millimeters	Millimeters tomography	Low (HPGe > NaI)	None	Intermediate to long, depending on # of spots	Simple	Aperture / collimator; detector efficiency characterization
Active coincidence neutron detection	🟨 ± <1.0 wt%	🟥 None	Centimeters	Low	None	Fast	Simple	Method to achieve spatial resolution

a. Accuracy and spatial resolution of systems as reported in the literature or online; the specifics for individual instruments may vary from these values.
b. Costs are estimated as low: $0–$50,000; medium: $50,000–$150,000; high: $150,000+.
c. Measurement times are estimated as fast: within a couple of hours; intermediate: approximately a day; slow: multiple days.
d. Operational complexity is estimated as simple: easily operated by nonexpert personnel, with little or no maintenance required; intermediate: easily operated by nonexpert personnel with some training, and some regular maintenance required; complex: substantial personnel training required to operate instrument and perform regular maintenance.

3.1 Technical Benchmarks

3.1.1 Accuracy of ^{235}U Concentration Measurement

It is extremely important to accurately measure the ^{235}U concentration, as a fraction of total U content, because it is this value that qualifies the fuel as LEU for regulatory purposes. Efficiency of the fuel elements in high-power reactors increases exponentially as the ^{235}U enrichment increases, and thus the target enrichment value is 19.75 ± 0.2 wt% ^{235}U, just below the designation of 20 wt% ^{235}U for HEU. Measurement of the ^{235}U enrichment with techniques that result in larger uncertainties can cause the material to be designated HEU. Measurement accuracy of ±0.1–0.2 wt% ^{235}U is challenging to achieve. The mass spectrometry-based methods (TIMS, ICP-MS, LA-ICP-MS) can achieve this accuracy (with improved accuracy for multi-collector setups), while gamma spectroscopy can reach it using the enrichment meter method. In the particular application of gamma spectroscopy to U-Mo alloys, the enrichment meter method will only produce accurate results if very well-characterized reference standards that contain precise quantities of ^{235}U are produced as U-Mo alloys. The National Institute of Standards and Technology (NIST)-traceable U oxide reference standards that can be purchased commercially will not be sufficient for this application, since both the ^{235}U density and the gamma-ray attenuation will differ in the oxide from that in the metallic U-Mo alloy. Analyzing the gamma-ray spectrum without employing reference standards, but instead estimating the detector efficiency, may not be able to achieve the desired accuracy. Autoradiography has no capability to quantify ^{235}U concentration in such a precise manner.

3.1.2 Spatial Resolution of ^{235}U Distribution

The ultimate goal of this characterization effort is to determine the spatial distribution of ^{235}U within the U-Mo alloy. This measurement will provide feedback to improve the casting process; once the process is optimized, the measurement will provide quality assurance that the ^{235}U distribution is acceptably uniform within the alloy. While measuring enrichment (Section 3.1.1) has received much attention and technological development, there has been little need before now to characterize the ^{235}U spatial distribution within a solid material. ICP-MS and TIMS are capable of providing larger-scale spatial distribution information, but at the expense of destructive testing: each area to be tested must be drilled, and a shaving measured by ICP-MS or TIMS. This methodology using ICP-MS is already being applied to the cast U-Mo plates at Y-12 with approximately centimeter spatial resolution. Material loss might be minimized, and spatial resolution improved, by drilling smaller sampling holes and efficiently collecting the shavings for analysis, but destructive sampling cannot be entirely avoided. Spatial resolution is likely limited to about 1 mm. Laser ablation-ICP-MS has distinct advantages in terms of spatial resolution. The spatial resolution is determined by the spot size of the laser light incident on the surface, which is typically 10–100 μm. LA-ICP-MS is a minimally destructive technique that removes only nanograms to micrograms of material during each pulse, creating a crater on the surface that is only a few nanometers deep. Autoradiography offers similar spatial resolution, with typical pixel sizes of 50–100 μm. Gamma spectroscopy does not typically offer inherent spatial resolution finer than the area of the detector, which is at least several square inches. The development of appropriate apertures and/or collimation, however, has the potential to reduce the detector FOV to the millimeter scale.

3.1.3 Ability to Meet Accuracy and Spatial Distribution Requirements

The ideal technology solution would be able to quantitatively measure the ^{235}U concentration with a high degree of accuracy and with fine spatial resolution. Technologies that cannot meet either requirement can be discarded without further consideration. However, technologies that can perform one task but not the other may still be useful as a portion of a combined approach, so the criterion was split into two components: the ability to meet the ^{235}U quantification requirements, and the ability to meet the spatial distribution requirements. The ^{235}U quantification requirement has been identified as ±0.2 wt% ^{235}U (as a fraction of total U) in the as-cast plates, and as ±0.1 wt% ^{235}U in the final rolled foils. The spatial resolution requirements have been estimated to be on the order of millimeters in the as-cast plates, and microns for the final rolled foils. For the purposes of this assessment, the spatial distribution requirement for the as-cast plates, which will be evaluated at the manufacturing facility, will be the criterion that the techniques are evaluated against. In Table 1, the ability of a given technology to meet these requirements is indicated by shading in the "Accuracy" and "Spatial Resolution" columns: red = does not meet criterion; yellow = possibly meets criterion; green = expected to meet criterion. All technologies except autoradiography meet one or both of the criteria at the yellow or green level.

3.1.4 Depth of Analysis

The ^{235}U may not be uniformly distributed across the cast plate, and likewise the distribution may be nonuniform within the depth of the plate (0.2" / 0.5 cm). TIMS and ICP-MS rely on shavings from test holes drilled in the plate, and these shavings are expected to represent a cross section of the plate depth. Analysis of these shavings will provide information averaged over the plate depth. Careful selection of sampling locations from various depths within the plate can impart depth information on the millimeter scale. In contrast, LA-ICP-MS samples only a few nanometers of the surface of the sample, and can be classified as a surface-sensitive technique. In theory, multiple laser pulses in the same location can provide depth-resolved composition data, but so little material is removed during each laser pulse that it becomes impractical to perform a depth profile measurement in a reasonable amount of time. Surface abrasion or chemical etching between measurements to remove material on a more macroscopic scale might be considered to provide gross depth information. Analysis of cross sections is another possibility. By its nature, gamma spectroscopy provides information from a significant depth of the material (likely millimeters), but in the form of total gamma-ray intensity with no depth-dependent information. The same is true for autoradiography. Gamma-ray tomography is possible, in which 3D information is extracted by modeling a series of 2D images collected at different angles or orientations (Hansen 2007). The spatial resolution of such a measurement would likely be on the order of a few millimeters (Svard et al. 2015).

3.2 Operational Benchmarks

3.2.1 Cost

The technology(ies) chosen to provide rapid feedback to the casting process will ideally be purchased or moved to the casting facility (Y-12). The cost of these instruments is a consideration in their adoption. In the absence of concrete pricing information, the cost has been estimated as "low" ($0–$50,000), "medium" ($50,000–$150,000), and "high" ($150,000+). Some technologies may prove useful for benchmarking the industrial process deployed, even if they are cost-prohibitive to set up at Y-12, so it should be noted that all technologies are available in some form at Pacific Northwest National Laboratory (PNNL); LA-ICP-MS is also available at Los Alamos National Laboratory (LANL).

3.2.2 Sample Preparation Required

It is preferable to measure the U-Mo as-cast plate or rolled foil directly, without any sample preparation or adverse effects on the material. Autoradiography, gamma spectroscopy, and LA-ICP-MS all can be executed with no sample preparation. Autoradiography and gamma spectroscopy are truly nondestructive techniques, while LA-ICP-MS is only minimally destructive, removing a few nanometers of surface material. In contrast, ICP-MS and TIMS are destructive techniques that require drilling or another method to collect material for analysis. For ICP-MS, the material to be analyzed is dissolved in acid. TIMS requires the element of interest to be separated from all other elements at high purity. This is a time-consuming process that must be optimized for the U-Mo system.

3.2.3 Measurement Time

Some techniques can produce data much faster than others. Measurement and subsequent data processing speed is not a primary concern for this project, since the technique will not be placed in the production line as a metrology tool, but it is still a factor to be considered. Autoradiography is expected to have the fastest measurement and processing time by far of any of the explored methods. Exposure of the phosphor detector to the U-Mo plate will require only seconds (the optimum exposure time produces a high-contrast image without saturating pixels), and read-out only requires an hour or two. Mapping with gamma spectroscopy cannot be accomplished with such a quick exposure. Instead, the spectrometer will collect data from a single region for several minutes (or possibly an hour or more) to achieve sufficient counting statistics before moving to the next location. Working with a 1 mm FOV on an as-cast U-Mo plate that measures 8" × 9" (~203 mm × ~228 mm), 46,284 areas would need to be measured to generate a map of the entire plate. This is likely a prohibitively large number of data points, and a sparser set of measurement locations would need to be determined. A similar situation arises for LA-ICP-MS, which has a spot size two orders of magnitude smaller than the estimated size for gamma spectroscopy, precluding analysis of the entire plate. Additionally, most LA-ICP-MS systems are not equipped to handle samples as large as the plate. Instead, analysis of disperse points on a smaller sample are feasible. With no time required to execute sample preparation, the LA-ICP-MS measurements should be fast (an hour or two) or intermediate (approximately a day), depending on how many spots are analyzed. In contrast, the fairly extensive sample generation (drilling) and preparation procedures required for ICP-MS increase the processing time; it may require several days to prepare and measure a series of samples collected from different locations on the cast plate. The necessity of separating the element of interest from other elements in the material encumbers TIMS with the longest processing times of any technique discussed. In addition, the low ionization efficiency of the evaporation process requires long measurement times to acquire sufficient counting statistics. TIMS is typically considered a low-throughput technique.

3.2.4 Operational Complexity

Any instrument that is deployed at the processing facility (Y-12) will be operated and maintained by nonexpert personnel. Gamma-ray spectrometers exhibit what is likely the lowest operational complexity. These spectrometers are commercially available turnkey systems that operate in a simple manner through a touch-screen or computer interface. They do not require regular maintenance. The equipment for autoradiography is likely to exhibit similar ease of use. The systems that incorporate ICP-MS, however, are more complex. High resolution ICP-MS systems are available commercially and are relatively straightforward to use. However, increased maintenance and troubleshooting compared to the gamma spectrometers is expected, rendering the complexity of ICP-MS “intermediate.” For LA-ICP-MS, in addition to operation and maintenance of the ICP and MS units, the nanosecond or femtosecond laser will require regular maintenance and alignment. Any gas chromatograph or other elemental separation technique used with TIMS will also require operation and maintenance. For these reasons, both LA-ICP-MS and TIMS are classified as “complex.”

3.2.5 Development

Lastly, not all the techniques discussed are available as COTS systems in the desired configuration for the purpose of measuring ^{235}U content and spatial distribution. The level of development required to produce a functioning system appropriate for this use needs to be taken into consideration. The only system that does not require any development is ICP-MS; Y-12 is already using this technique to analyze shavings from U-Mo plates. Likewise, LA-ICP-MS requires no development for this application. Spatially resolved gamma spectroscopy requires the development of apertures and/or collimators to reduce the detector FOV to the millimeter scale. These should not be technically challenging to design. For the enrichment meter method, well-characterized reference standards of U-Mo with very precise ^{235}U concentration are required; a path to obtaining and qualifying these standards is not immediately clear. For standardless gamma spectroscopy measurements, an accurate and effective method to characterize the detector efficiency is required, but there is no agreement in the literature as to the best approach to use. Autoradiography requires the most development of any technique discussed here, since it offers no inherent isotopic sensitivity in the ^{235}U/^{238}U system.

4.0 Recommendations

Based on the accuracy and spatial resolution parameters summarized in Table 1, both LA-ICP-MS and gamma spectroscopy using the enrichment meter method meet the requirements to quantify ^{235}U enrichment in as-cast U-Mo plates. From the other parameters in Table 1, LA-ICP-MS is not ideally suited for deployment to Y-12. A commercial system is both expensive and complex. The recommendation for LA-ICP-MS is to utilize the existing systems at LANL and PNNL as benchmarking tools to validate the chosen technology deployed to Y-12. In addition, LA-ICP-MS at LANL and/or PNNL will be necessary to confirm the ^{235}U distribution homogeneity of the final rolled foil fuel elements, where micron-scale spatial resolution is required.

For near-real-time nondestructive analysis of the as-cast U-Mo plates at Y-12, gamma spectroscopy is recommended. This necessitates the acquisition of a HPGe gamma spectrometer. These are common instruments, and a new purchase may not be necessary. The development of a collimator/aperture system to achieve millimeter or better spatial resolution will be straightforward. Once the measurement time required to obtain satisfactory counting statistics is determined, a statistical analysis can be undertaken to determine the most efficient sampling scheme (number and spacing of sampled areas). A jig will need to be fabricated to hold the spectrometer and plate in a defined geometry (distance between plate and detector, in parallel arrangement); the jig must facilitate movement of the spectrometer and/or plate to measure the specified areas. A schematic of one possible setup is shown in Figure 1.

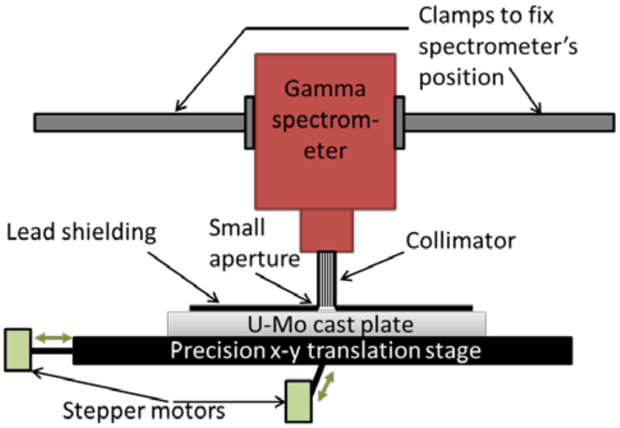

Figure 1. Schematic of a Jig to Map the U-Mo Cast Plate (gray) with Gamma Spectroscopy (red). The spectrometer is held in fixed position, and the U-Mo plate is translated in x,y.

To accurately quantify the ^{235}U concentration in each sampled area using the enrichment meter method, ^{235}U standards are required. As discussed in Section 3.1.1, commercially available ^{235}U standards will not be sufficient for this purpose since they do not possess the same enrichment, density, geometry, or material composition as the plates to be measured. Instead, it will be necessary to either purchase a custom-made calibration standard (may be NIST-traceable), or fabricate standards in-house by arc-melting and rolling buttons. Standards fabricated in-house then require precise measurements to establish the exact ^{235}U content.

Gamma-ray tomography has potential to reveal the ^{235}U distribution within the depth of the as-cast plate with a resolution of a few millimeters. However, this technique is not recommended here as a standard component of the gamma spectroscopy measurements because it is nontrivial to set up, and the best tomographic data would be obtained on a slice of the as-cast plate, not the entire plate, making this a destructive technique. However, it could be useful for certain benchmarking tasks, particularly if a facility could be identified that already possessed the capability.

ICP-MS and TIMS are not recommended because they are destructive measurements, but, as with the tomography, they may be employed on a limited basis for benchmarking. The complexity of setting up a neutron detection scheme with an active neutron source incident on the sample and multiple neutron detectors is too complex, given the lack of spatial information and relatively poor ^{235}U enrichment accuracy that can be expected from the measurements, and thus neutron-based techniques are not recommended.

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