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AT-LINE PROCESS MONITORING FOR NUCLEAR SAFEGUARDS

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ABSTRACT

The long-term goal for international safeguards is to transition to unattended, near real-time verification. To support this goal, the International Atomic Energy Agency (IAEA) has requested new technologies for process monitoring and near real-time accountancy. As has been shown in other industries, at-line process monitoring is a method by which the international community could meet the IAEA's stated goals by automating in-process sampling and analysis. By integrating automation, high-throughput sampling and analysis, advanced analytics and machine learning, and cross-validation using multiple different near real-time techniques, it should be possible to achieve unattended near real-time accountancy with overall uncertainty levels comparable to, or better than, current off-line approaches. This paper endeavors to properly define at-line process monitoring as applied to safeguards and will discuss researchers' findings on the potential role of automated sampling-based measurements to meet safeguards near real-time accountancy and process monitoring needs. Furthermore, the authors hope this paper initiates a conversation on the values of at-line process monitoring as a grander part of the international safeguards regime.

INTRODUCTION

Traditional nuclear safeguards approaches require a significant amount of costly and time-consuming manual sampling, sample processing, and destructive analysis in a laboratory. Although there has been recent progress in simplifying and automating some off-line sample preparation and analysis operations, there is a growing interest in reducing the burden of laboratory analysis by using near real-time process monitoring (PM) data to draw safeguards conclusions. Towards this end many research programs are focused on the development of in-situ and in-line techniques for safeguards monitoring. However, most groups have not considered that in-process sampling and analysis (i.e., at-line process monitoring) could be deployed to meet the safeguards goals outlined by the IAEA.

At-line PM has been used for decades in the pharmaceutical, biotechnology, and chemical industries to achieve near real-time analysis using analytical techniques that are either too complex or not robust enough for in-situ, in-line, or on-line use. In these industries, off-line techniques are often streamlined and automated to become at-line techniques.[1] The transition from off-line to at-line techniques enables more rapid feedback and can facilitate the use of high-throughput sample generation and analysis, which can improve measurement precision through the averaging-out of random sampling and measurement errors.[2] Due to the need for more timely diversion detection, the desire to move away from costly manual techniques, and the harsh environment of most nuclear safeguards applications, at-line PM should be considered for near real-time accountancy applications.

This paper discusses the potential role of automated sampling-based measurements to meet safeguards near real-time accountancy and PM needs; provides examples of techniques that could be deployed for at-line monitoring in molten salt facilities or gas centrifuge enrichment plants; and discusses some of the current roadblocks to the implementation of at-line approaches. Since at-line techniques have not been part of the nuclear safeguards discussion to date, one key roadblock is basic

stakeholder awareness. This report aims to increase awareness of the at-line approach and includes recommended standardized PM terminology to help improve communication among stakeholders.

PROCESS MONITORING TERMINOLOGY

There are no universally accepted definitions for chemical processing monitoring terms. Within the nuclear field, the definition of the simple term "process monitoring" varies significantly. One safeguards workshop report defined PM as being "continuous and unattended."[3] Coble et al. stated that "process monitoring can include any bulk measurement within a facility measured in-situ, that is, non-sampling measurement techniques."[4] However, the broader chemical industry does not exclude non-continuous analysis or sampling-based methods from their definitions of PM. Cipiti specified limited-use definitions of PM as "any non-elemental quantification measurement"[5] and "any non-nuclear bulk measurement in a facility, including mass, density, level, current, voltage, temperature, etc., or an on-line measurement that can provide more continuous data of process streams including spectroscopy, pH monitors, flowmeters, off-gas monitors, etc.".[6] While these definitions are useful for providing clarity within a single report, they could cause confusion if improperly cited. A broader definition of PM—along with more specific terminology to characterize types of PM—would be useful to facilitate communication between stakeholders on a broader scale.

Companies that sell PM instrumentation to the chemical and biotechnology industries generally categorize PM tools into in-line/in-situ, on-line, at-line, and off-line, depending on where in the process the analysis takes place (see Figure 1).[7] With the exception of at-line, these terms are also used in the nuclear field where their usage varies widely. It is generally accepted that off-line analysis involves the removal of discrete samples from a process and sample transport to a separate location for analysis. Techniques that do not fit this definition are often grouped together as "on-line" techniques. However, this broad categorization is not ideal for communication among stakeholders because it covers a wide variety of tools and techniques that have vastly different concept of operations (CONOPS), costs, and safeguards implications.

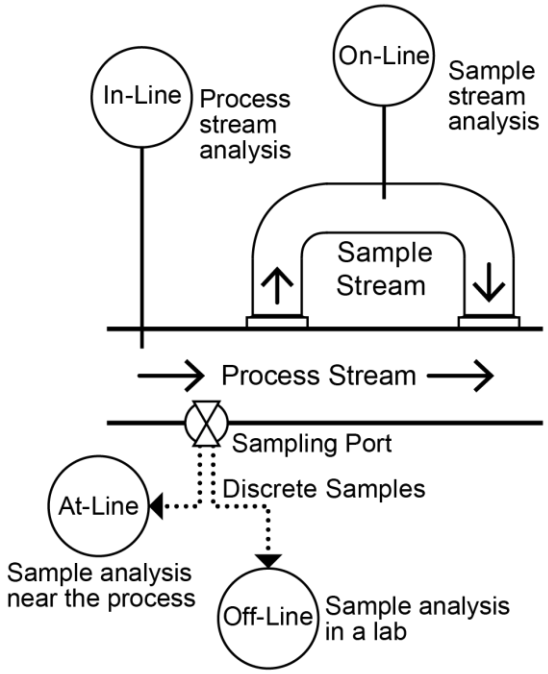

Figure 1. PM terminology based on process location.

PM is broadly defined below and subdivided into categories with simple definitions based on the location of the material being analyzed. These definitions generally align with the accepted terminology in the chemical and biotechnology industries. In the broader chemical industry, the at-line versus off-line distinction is determined by whether the sample is analyzed on the plant floor or in a separate analytical laboratory. However, this boundary can be less clear in nuclear safeguards applications involving hot cell infrastructure. Therefore, an additional distinction based on sample custody is proposed here. Techniques are classified as "in-process" if the samples remain in "process custody" or as "off-line" if they are transferred into "human custody" (refer to Table 1). This distinction has important implications for at-line safeguards monitoring because techniques that fall under the in-process categorization can be deployed as unattended monitoring systems.

Table 1. Process Monitoring for Nuclear Safeguards

In-process Analysis			Off-line Analysis		
In-line/In-situ	On-line	Unattended At-line	Manual At-line	On-site Lab	Off-site Lab

Recommended Process Monitoring Terminology and Definitions

Process Monitoring - The measurement of any process variable including, but not limited to, state variables (temperature, mass, volume, etc.), bulk properties (level, density, etc.), chemical or isotopic composition, speciation, and spectroscopic or radiation signatures. Includes in-line, in-situ, on-line, at-line, and off-line methods. Can be fully automated, partially automated, or manual. Can be continuous or intermittent monitoring. Can be implemented to provide feedback for process control or for other purposes (i.e., safeguards measurements)

In-situ/In-line Process Monitoring - Analysis of material that is in a process vessel (in-situ) or in a process pipe (in-line). Instrument can be portable or fixed. The technique can be invasive or non-invasive depending on whether the detector is exposed to the process fluid. (For example, an electrochemical probe would be an invasive technique, while gamma measurements through a pipe wall would be non-invasive.)

On-line Process Monitoring - Analysis of process material within a sample stream, sampling loop, or split stream. Instrument can be portable or fixed. Technique can be invasive or non-invasive

At-line Process Monitoring - Removal of discrete samples from a process followed by sample handling, sample processing, and sample analysis near the process. Can be fully automated to support unattended process monitoring or may include manual steps

Unattended At-line Process Monitoring - Fully automated sampling, sample handling, sample processing, and sample analysis. Instrument can be portable or fixed

Manual At-line Process Monitoring - At-line process monitoring with at least one manual sample handling, sample processing, or sample analysis operation

In-process Analysis - Analysis of a process variable where no material is removed from process custody (i.e., material remains in a process vessel, process line, sample line, or in the custody of an automated sample handling and analysis system)

Off-line Analysis - Analysis of samples in human custody. Samples may be physically transferred to human custody (e.g., for analysis in an analytical laboratory) or might be operationally transferred (e.g., samples remain in the hot cell infrastructure but are processed by one or more manual steps)

Portable Instrument - Movable instrument that does not require laboratory infrastructure to operate. Can be robot- or human-portable. Can be used for in-process or off-line monitoring

Note that the terms "in-field" and "field-deployable" were not included above. These terms hint at portability and environmental analysis and are commonly used in nuclear forensics and by inspectors to distinguish tools that do not require laboratory infrastructure. These terms are sometimes used interchangeably with the term "on-site"[8,9] but this usage is not ideal for providing clarity in CONOPS discussions. For example, a handheld detector and an on-site mass spectrometry system might both be labeled as field-deployable under this usage yet would have vastly different CONOPS. Therefor it would be beneficial to avoid the terms "field-deployable" and "in-field" when describing PM applications. More specific terms such as "in-process" and "process-deployable" can be used to describe techniques that are suitable for in-line, on-line, and automated at-line deployment; and terms such as "on-site lab" and "inspector portable" can be used when describing field-deployable instrumentation that is intended for off-line use.

HIGH-THROUGHPUT ANALYSIS AND ADVANCED INTEGRATION APPROACHES

Material accountancy in molten salt reprocessing and liquid-fueled molten salt reactor facilities is challenging due to high temperatures, corrosivity, complex fluid mixtures, and complex material flow and fluxes.[6,10] In order to distinguish between normal operational variations and diversion events, these facilities will require a sophisticated monitoring approach that relies on PM to address gaps left by traditional safeguards approaches.[11] Several new PM tools are under development to provide high-fidelity material tracking measurements at key points in facilities with the overarching goal of developing a system of unattended measurements that can be integrated to supplement or replace traditional manual sampling and off-line analysis.

One PM approach currently under development is at-line high-throughput sampling and analysis (HTSA). HTSA is used in a variety of industries because this approach can improve the precision and limits of detection for existing analytical techniques simply by averaging out the random statistical error associated with sampling and measurement. This effect is represented by Equation 1, which describes the random variability associated with a set of measurements.

$$CI = x \pm t * \frac{s}{N} \quad (1)$$

Here CI represents the confidence interval for the set of measurements, x is the mean value of the samples measured, t is the distribution representing the sample population, s is the standard deviation of the measured values, and N is the number of samples. High-throughput sampling and analysis (i.e., high N) can narrow the confidence interval compared to traditional safeguards measurements where only one or a few samples are analyzed. While high-throughput sample characterization has the potential to significantly improve analysis precision for a variety of analytical techniques, there are currently very few methods available for the high-throughput analysis of nuclear samples. The extensive sample preparation required for many gold standard measurement techniques is a barrier to high-throughput sample characterization. However, as described in the next section, there are existing analytical techniques that are amenable to automation and in-process deployment as unattended, at-line HTSA PM tools.

Given the improvements in precision that can be achieved by high-throughput analysis, it may be possible to achieve the target uncertainty levels requested by the IAEA using HTSA versions of analytical techniques that require little or no sample preparation. While the individual measurements may not be able to achieve the measurement uncertainty levels of gold standard techniques,

high-throughput sampling could achieve better overall uncertainty levels by reducing the errors associated with sampling. Furthermore, at-line and other near real-time techniques can be combined to improve diversion detection using an integrated approach. Researchers are currently developing capabilities to "extract knowledge, make use of correlations, and quantify uncertainty in an integrated manner" to achieve higher levels of facility operational awareness than can be achieved with current non-integrated approaches that do not take full advantage of all available data.[12] For example, gamma spectroscopy can determine plutonium content with a relatively high measurement uncertainty of 5-10%, which is too high for reliable diversion detection. However, by integrating multiple PM data streams using advanced approaches like machine learning, gamma measurements could provide a valuable contribution to a diversion detection mechanism.[13] Other PM data to be integrated with this approach will likely include electrochemical measurements and sampling-based measurements.[2] Ideally, sampling-based measurements will be performed in near real-time by implementing unattended at-line PM technologies such as the example techniques described below.

EXAMPLES OF TECHNIQUES SUITABLE FOR AT-LINE PROCESS MONITORING

At-line Monitoring for Uranium Hexafluoride Process Gas Enrichment Verification

The IAEA's current safeguards enrichment verification approach for gas centrifuge enrichment plants (GCEPs) combines sampling with destructive analysis at IAEA Nuclear Material Laboratories using sophisticated benchmark mass spectrometry techniques that are not suitable for on-site deployment.[8] To improve the timeliness of enrichment verification, a variety of sampling-based mass spectrometry and optical techniques (summarized elsewhere[8,9]) are under consideration for on-site deployment. However, most on-site techniques described to date require either complex sample preparation or manual instrument operation by a skilled worker. To further improve the timeliness of analysis and to remove the costs and risks associated with manual operations, the On-line Enrichment Monitor (OLEM) was developed.[14] This instrument measures the ^{235}U gamma signal from the exterior of a process pipe and estimates the enrichment level using temperature and pressure information provided by the plant operator's in-line instruments. Some key limitations of the OLEM are its inability to directly measure ^{238}U and ^{234}U, its reliance on operator instruments, and its reliance on a variety of assumptions. Because of the limitations of these existing technologies, the IAEA has requested further investments by member states to develop new technologies for the near real-time detection of highly enriched uranium (HEU) production in low enriched uranium (LEU) GCEPs. Guidance documents state that the IAEA is specifically interested in low-cost technologies that can be fully automated for unattended analysis.[15] Three technologies developed at U.S. National Laboratories in response to this request include the laser-induced spectrochemical assay for uranium enrichment (LISA-UE), high performance infrared (HPIR) spectroscopy, and in-field alpha spectrometry (IFAS). These techniques are currently targeted for manual at-line deployment; however, as discussed below, each of these techniques has potential to be deployed as automated at-line techniques to fulfill the IAEAs request for unattended monitoring capabilities.

LISA-UE functions by creating laser induced plasma in a gaseous UF_6 sample contained in a sampling chamber. The isotopic shift in emission wavelength provides the basis for enrichment measurement.[8] A benefit of the LISA-UE method is that the enrichment level of UF_6 can be obtained directly from the measured spectra without the need to use a calibration standard. HPIR spectroscopy also uses gas-phase UF_6 samples but relies on the isotopic peak shift for infrared absorption at 1163 cm^{-1} . The HPIR method has been demonstrated to meet IAEA target values for non-destructive analysis and could likely be operated to meet destructive analysis criteria with longer analysis times.[16] The HPIR system was originally envisioned as being part of a sampling loop ("on-line" deployment). However,

stakeholder engagement revealed significant challenges to this approach including operator preferences, hold-up issues, and development, installation, operation, and maintenance costs. In light of these challenges, at-line deployment could be a better path forward for this technology. Because both the HPIR and LISA-UE techniques are robust, provide rapid analysis, and can utilize gas phase samples with no sample preparation, they are amenable to being fully automated. The measurement operations that follow sample delivery to the instruments could be automated with very little R&D investment, while full automation of the UF_6 sampling operations would require a larger investment. However, due to the low radiation field in GCEPs relative to other fuel cycle facilities, there is the potential to implement a wide variety of sophisticated automation solutions. Furthermore, expertise from the laboratory automation and industrial automation sectors could be leveraged to accelerate the process. For example, it's likely that much of the automation engineering could be outsourced to vendors that provide commercial automation solutions.

A third enrichment verification method that may be suitable for unattended at-line deployment is the Single-use Destructive Assay Sampler (SUDA)[17] with In-field Alpha Spectrometry (IFAS)[18]. The SUDA sampler is small and robust, consisting of a coupon wafer with a deposited film of absorptive zeolite. The coupon is connected to a sample port and exposed to gaseous UF_6, which is converted to UO_2F_2 within a few minutes. Researchers developed the SUDA sampler to address issues related to transportation, chain of custody, and analysis timeliness for destructive analysis of UF_6 samples collected at GCEPs. In response to an IAEA request for an alpha spectrometric method suitable for in-field enrichment verification[19], the IFAS alpha spectrometer system is being developed to interface with the SUDA sampler to allow quick turnaround measurements of UF_6 process gas enrichment. This approach avoids the need for traditional alpha spectrometry sample preparation, which is rigorous and time-consuming. Because alpha spectrometry can be used to directly measure the three uranium isotopes of interest and does not rely on any of the assumptions required by the OLEM, it is expected to be a useful tool for validating OLEM measurements. The IFAS system was developed for near-term manual at-line deployment and is to be either inspector-portable or stored on-site in a secure IAEA instrument cabinet. However, once the technique is proven as a manual at-line approach, it should be possible to quickly adapt the system for unattended operation by leveraging resources from the automation industry.

For automated sampling and analysis by any of the three methods, the samples may be collected and then transported to an instrument at a fixed location (e.g., by an airlift system), or the instrument itself could be transported to the sampling ports (e.g., by a robotic cart system). Either of these approaches would eliminate human error and other safeguards risks associated with manual sampling and human custody of samples. One key challenge would be the development of mechanisms to ensure the security of the unattended sample transport or instrument transport operations at the GCEPs (where, unlike many other fuel cycle facilities, the temperatures and radiation fields do not preclude human access to the process). While the up-front development and implementation costs would be significantly greater for automated versus manual at-line deployment, the authors believe that full automation should be considered because the unattended at-line deployment of one of these techniques would drastically improve the timeliness of OLEM measurement verification at GCEPs while eliminating the costs and risks associated with manual operations.

Microcalorimeter Techniques for At-line Salt Characterization

Fully automated sampling and sample analysis will be more challenging in molten salt applications versus GCEPs due to the extreme radiation and temperatures. However, the development of in-situ and on-line methods is also challenging for the same reason and the removal of samples for off-line

analysis introduces a safeguards risk that could be avoided with the use of at-line techniques. Furthermore, by developing salt sampling technologies to enable at-line analysis, a variety of high-precision techniques could be deployed for near real-time unattended analysis at pyroprocessing and molten salt reactor facilities. One example is the Spectrometer Optimized for Facility Integrated Applications (SOFIA), which is a cryogenic microcalorimeter that provides unprecedented energy resolution for x-ray and gamma-ray spectroscopy.[20] Microcalorimeter detectors work by converting the energy of an incident photon to heat in an absorber and then measuring the temperature rise in the absorber, which is proportional to the energy of the deposited photon. The IAEA Nuclear Material Laboratory has expressed interest in using these techniques for analytical laboratory characterization of safeguards samples; however, recent innovations now make it possible to take this tool out of the laboratory and deploy it for in-process safeguards measurements. Specifically, modern refrigeration technology has enabled microcalorimeter systems that operate without the need for liquid cryogens and with utility requirements that are no more demanding than a window air conditioning unit, thus making this tool suitable for operation in nearly any facility. Additionally, new instrument geometry makes it amenable to automated (and high throughput) sample analysis because samples of various sizes are simply positioned next to the unit's cryostat for measurement. Finally, SOFIA can be easily relocated to measure samples that are difficult to move or handle. With its unprecedented energy resolution, the latest deployability innovations, and the ability to easily integrate samples with the SOFIA unit, this instrument is a top candidate to support at-line PM applications.

Microcalorimeter gamma-spectroscopy can provide isotopic and elemental information with energy resolutions that are up to 10 times better than conventional high-purity germanium detectors.[20] Improved energy resolution helps to separate closely spaced spectral features and to detect weak spectral features against strong backgrounds like those encountered from samples that contain fission products. This capability is ideal for measuring actinide content in complex multi-actinide samples with fission products (e.g., electrochemical fuel reprocessing samples). Ultra-high resolution microcalorimeter gamma spectroscopy is already being used to inform safeguards models and to explore signatures of spent fuel and reprocessing. In the future, microcalorimeter gamma spectroscopy could be deployed in various nuclear processes to provide completely passive nondestructive isotopic analysis with sufficient precision and accuracy to reduce requirements for costly chemical separations and mass spectrometry analysis. SOFIA could be deployed for at-line, non-invasive on-line, or non-invasive in-line safeguards PM. For very high radiation applications, sampling and sample transport away from the extreme radiation zones would be required to enable in-process microcalorimetry measurements.

Salt sampling and sample handling are key remaining challenges for at-line PM of molten salt facilities[7,21,22] because they will require significant additional investment to develop and performance test tools that are robust enough for continuous remote operation in extreme temperature and radiation conditions. However, integrated sampling and microcalorimeter analysis capabilities would be a powerful tool for enabling rapid, automated, and unattended at-line safeguard measurements with laboratory-grade precision and accuracy. Additionally, a single salt sampling unit could be integrated with a variety of at-line analysis techniques such as laser induced breakdown spectroscopy, x-ray fluorescence, radiometric techniques, etc. These data streams could be used both for material tracking/verification measurements and for process control/optimization purposes. By reducing normal process variation through improved process control, it will be easier to detect off-normal process conditions of security concern. Therefore, this dual-purpose at-line monitoring approach would maximize the diversion detection capabilities of an advanced integration safeguards system.

TIME AND COST FOR DIFFERENT PROCESS MONITORING APPROACHES

The successful deployment of at-line PM tools in other industries indicates that this approach could have significant potential benefits for safeguards at a variety of fuel cycle facilities. However, when making safeguards decisions, these benefits must be weighed against the remaining technical challenges, costs, and timeline to deployment. Figure 2 illustrates the trade-offs for PM tool deployment options. The right side of the plot represents manual sampling coupled with off-site analytical laboratory techniques, which constitute the existing gold standard techniques. Aside from some efficiency improvements, these techniques will require very little R&D investment going forward. However, the measurement turnaround time can be several weeks, and the cost per measurement is very high due to the sample transport and skilled labor requirements. To the left of the off-site techniques are the on-site analytical laboratory techniques. Many of these still require additional R&D investment, however, they all have the benefit of quicker turnaround times and slightly lower costs per measurement than off-site approaches.

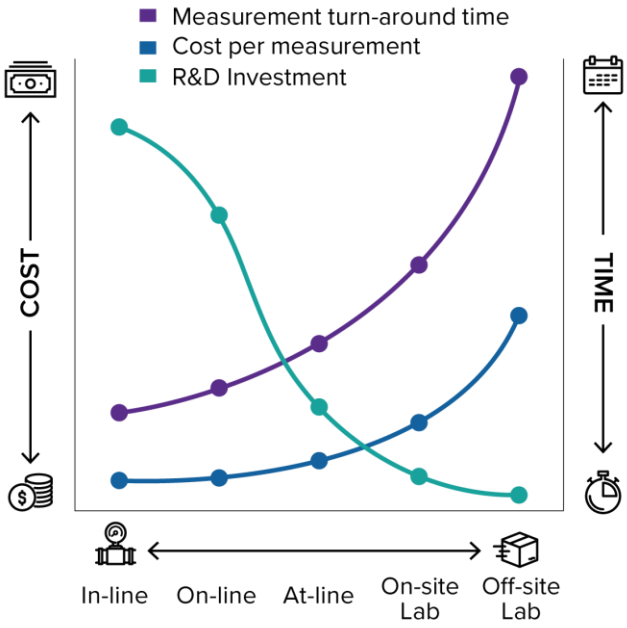

Figure 2. Qualitative analysis of time and cost for different PM tool deployment approaches.

On the left side of the plot in Figure 2 are invasive in-line techniques. All invasive in-line techniques (including the well-developed technologies) will have high development costs related to process integration. This is because (1) the installation and maintenance of these instruments can be costly and disruptive to the process, (2) the footprint of each sensor in the process must be considered, and (3) the reliability of each sensor must be demonstrated under appropriate process conditions. However, in-line measurements are real-time or near real-time and the cost per measurement is negligible relative to off-line analysis. To the right of the in-line techniques are the invasive on-line techniques. Like invasive in-line measurements, invasive on-line measurement techniques have high R&D costs related to process integration and reliability testing. However, on-line techniques have a smaller process footprint (generally a single sampling line), and maintenance operations are much simpler than for invasive in-line techniques. Furthermore, once sampling loop technology is available, the cost to deploy each individual on-line analytical technique will be less. In the middle of the plot are the at-line techniques. Fully automated sampling-based analysis techniques generally have not been considered for in-process safeguards monitoring. However, they could be a better alternative to

some techniques that are currently in use or under development as in-line, on-line, or off-line techniques. This is because, firstly, they will require the least instrument radiation hardness of all the in-process analytical techniques, which will enable the use of a wide variety of techniques that can currently only be used off-line. Secondly, automated at-line techniques could support high-throughput sample analysis to improve the accuracy and precision of existing techniques by eliminating human error and reducing random errors associated with measurements and sampling inhomogeneity. Thirdly, although enabling technologies (e.g., precision salt samplers) for at-line molten salt analysis are currently at low technology readiness levels, once these enabling technologies are deployed, the cost to implement each at-line analytical technique will be low due to lower temperatures and radiation fields, minimal process impact, and lower costs for maintenance operations. Finally, many at-line technologies could be adapted from current off-line methods quickly and at relatively low cost by leveraging existing automation technology and industry resources.

CONCLUSIONS

Next-generation nuclear safeguards will implement advanced integration of process modeling, process monitoring, and data analytics to achieve near real-time material accountancy. Due to the potential benefits listed here, at-line PM approaches should be considered for inclusion in this next generation safeguards paradigm. The final suite of monitoring tools for each process will be selected through an iterative development process with consideration for operator preferences, evolving regulatory requirements, costs, funding, and technology readiness. Some PM technologies (e.g., electrochemical sensors) are on track for near-term deployment as part of this next-generation safeguards approach. However, to fully meet safeguards requirements, these techniques will require validation with higher-precision methods and significant roadblocks still exist for the unattended in-process deployment of higher-precision techniques. These roadblocks include technology gaps, gaps in the regulatory framework, and disconnect between technology developers, potential end users, funding organizations, and policy makers. Until these issues are addressed, a large portion of safeguards monitoring will continue to be performed with outdated, untimely off-line analysis techniques.

To date, most reports on safeguards measurement techniques are focused on the accuracy and precision of the analytical techniques and there is no doubt that sponsors will continue to fund researchers to develop new and improved analytical techniques for nuclear safeguards measurements. However, the path forward for integrating these techniques into industrial processes is still unclear, particularly for molten salt facilities. In addition to technical gaps such as operations and maintenance tooling and salt sampling systems, there are uncertainties such as the issue of who will pay for and perform the extensive reliability and performance testing required to deploy new PM technologies in high-radiation remote-operation facilities. In order to address the gaps and realize the benefits of in-process monitoring methods as part of an integrated safeguards approach, discussions must take place among stakeholders to determine the best approach for integrating each process monitoring technology into the physical process, into the process operations, and into the integrated safeguards workflow. This paper aimed to help facilitate those discussions by providing recommended terminology for clarity and by outlining the potential benefits of underutilized automated sampling and analysis approaches.

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