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Assessment of Process Monitoring Techniques for Pyroprocessing Safeguards

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1. Introduction

Pyroprocessing is a technology, currently under development, to support recycling of spent nuclear fuel. Due to lack of input accountability and impracticality of flush-out [1], application of the safeguards approach based on Nuclear Material Accountancy (NMA) to pyroprocessing appears difficult. The limitations of NMA can be complemented by the use of Containment/Surveillance and Process Monitoring (PM) technologies. In particular, PM technologies can be used to inspect normal/off-normal operation with various data obtained from facility operations in real time to meet safeguards objectives.

To support the use of PM technologies for the purpose of pyroprocessing safeguards, this study aims at identifying technologies that could be useful for PM purposes and evaluating their applicability to a pyroprocessing facility.

This paper describes the development of the assessment criteria to evaluate the practicality of candidate technologies for PM based on a variety of requirements and considerations. By using the developed assessment criteria, application of technologies in the oxide reduction process was assessed as a test case example.

2. Methods and Results

In this section, the oxide reduction process is explained and candidate PM technologies are investigated. Also considerations for developing assessment criteria are introduced.

2.1 Listing of Possible PM Technologies for the Oxide Reduction Process

In the oxide reduction process, SF is reduced from the oxide metal form to the metal form by removing oxygen. Through this process, the volume of SF is reduced and high heat load fission products, such as Cs, Sr, and Ba, are dissolved out of the SF and accumulated in electrolyte. In this system, SF is loaded into a stainless steel basket coated with porous MgO membrane. The basket acts as a cathode, And Pt is used as an anode. The electrolyte is a 1 wt.% Li_2O-LiCl molten salt system. When electric current passes through the cathode, oxide metal is reduced to metal or dissolved out, in the chloride form, according to the reactivity of each element. On the surface of the anode, oxygen ions are oxidized to oxygen gas. The gas is removed from the system through dedicated tube(s) and collected outside of the reduction system [3].

If the process is operated under off-normal conditions, process monitoring becomes important since it can significantly influence the material accountancy of the whole process. Almost all processes are operated in Ar atmosphere including oxide reduction. In this regard, it is important to monitor the oxygen removal. It can reduce the possibility of corrosion in the whole process. Also, it enables process monitoring and system diagnosis through predicting reduction rate by measuring the amount of released oxygen.

It is rare, but the possibility exists that U and Pu can melt into molten salt, in chloride form, during abnormal process conditions. If the oxides are not completely reduced, they can remain in the cathode basket. The problem can occur in the NMA process because the actual throughput can be different from the estimated amount. Some techniques can be applied as PM technology to assure normal operating condition; oxygen sensors and flowmeters can monitor the reaction rate; “power supply”, “temperature”, “molten salt stirring speed”, “salt composition and concentration”, “electrometer, voltmeter and amperemeter” can directly monitor the operating condition. As an example, a detailed explanation of the usage of oxygen sensors and flowmeters is provided in 3.2.

2.2 Considerations for Developing Assessment Criteria

A systematic method is required for comparing different technologies in terms of readiness and application. The method should consider the performance of the technology in terms of safeguards (IAEA criteria) and operating conditions of pyroprocessing. Further discussions are given below in the consideration of technology readiness, operating environment, and applicability of the technology.

Technology Readiness Level (TRL); TRL was pioneered by the National Aeronautics and Space Administration (NASA) to manage the technology investment of the aerospace industry. Sadin at el. (1989) suggested an evaluation system for comparing the maturity of different technologies. The TRL is divided from Level 1 (basic principles observed and reported) to Level 9 (proven system through successful operation in system being evaluated) [4].

Operation environment in pyroprocessing; equipment for monitoring is installed inside a hot cell where the concentration of oxygen and moisture is maintained at an extremely low level (O_2, H_2O < 50 ppm) to prevent corrosion. However, the environment is still corrosive because of the presence of chloride salt in the process. Also, the equipment is exposed to high temperature and radiation. Because maintaining a consistent environment in the hot cell is an essential factor for a stable operation, the access for maintenance and repair of equipment is limited. The performance of equipment can be degraded and even operated in unstable conditions in this regard. Therefore, the installed equipment should function and maintain its integrity in the operation environment.

Applicability of a technology; process monitoring technology should not influence the process, and it should provide process data in real time, or near real time. The equipment should not obstruct the flow of the original process due to prolonged analysis time and/or space requirements.

Though PM technology is used to supplement NMA, if the candidate technology requires a composition analysis, the maximum permissible variance error and concentration should also be considered. The purpose of the composition analysis is to prove the proliferation resistance of the process. This is done by demonstrating that the expected error is less than SQ (the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded) within the conversion time (the time required to convert different forms of nuclear material into the metallic components of a nuclear explosive device) [2]. This criterion will be the minimum requirement for composition analysis technologies.

3. Result and Discussion

3.1 Assessment Criteria

Based on the considerations described in section 2.2, assessment criteria for PM technology are derived as follows.

1) Process interference or process material destruction; the technology should not interrupt the flow of a process. Though DA is known for its analytical precision, if the process is impeded during sampling or for other reasons, the technology can be rejected.
2) Possibility of real time detection; to ensure the proliferation resistance of the pyroprocessing, the real time detection is an indispensable factor. The candidate technology should detect or analyze any change (concentration or mass transfer) in real time or near real time.
3) Ease of installation; the candidate technology can be adapted without or with minor changes to the current pyroprocessing facility design.
4) Technology Readiness Level; this assessment criterion is a systematic and analytic metric that compares the maturity of heterogeneous technologies. Various candidate technologies which have different characteristic can be compared with this criterion.
5) Durability and life expectancy of equipment; pyroprocessing is operated in a severe environment which includes high temperature and radiation. This inevitably results in physical stress to the equipment. Because the equipment is located in a hot cell, a repair or replacement of equipment is challenging. In addition, if the equipment cost is expensive (e.g., spectrometer and counter), the cost of maintenance will be significantly high. Therefore, the candidate technology should have a high durability and long life expectancy.

When a candidate technology requires a composition analysis, there should be additional criteria. These criteria have to prove the proliferation resistance of the process, demonstrating that the expected error is less than SQ and have a defined conversion time.

6) Detection Limit; the technology should continuously detect certain elements with relatively low concentrations. The detection limit of the analytical equipment should be sufficiently low to adequately measure the elements of interest. If the detection limit is lower than the maximum permissible variance error and concentration, the technology should meet the safeguards requirements.
7) Degree of precision and sensitivity; this criterion is associated with maximum permissible variance error and concentration. By satisfying this criterion, these analytical technologies will meet the IAEA's safeguards requirement (SQ).
8) Possibility of selective analysis; certain species which is highly influential in non-proliferation should be selectively analyzed.
9) Generation of secondary waste; when the sample is prepared with certain pretreatment, the secondary waste should be avoided because there can be special nuclear material in the disposed sample that negatively influence in proliferation aspect.

3.2 Adapting these Assessment Criteria to the Oxide Reduction Process

The assessment criteria derived in section 3.1 were applied to candidate PM technologies in the oxide reduction process. The results are presented in Table I. As shown in Table I, most evaluated technologies were found to have high scores. The only exception was the analysis technology (concentration and composition of salt). In the table, a number is allocated to a technology according to its suitability in matching an assessment criterion. Where, '1' is allocated to a suitable technology, '0' is allocated to an unsuitable technology, and '0.5' applies to technologies that have not been deemed suitable or unsuitable. If a technology is not related to a criterion, the numeric designation is left blank. The total score, for an individual technology, is calculated by dividing 'the sum of the allocated numbers to that technology' by 'the number of determined criteria (except blanks in the table)'.

A flow meter is an important candidate PM technology. The flow rate of fluid can be detected and be used to estimate the degree of reaction that occurs on the anode. This is done by measuring the amount of oxygen gas generated when metal oxide is reduced to metal on the anode. The flow meter is installed where the off-gas exits the system [5]. Considering the principle of how a flow meter works, this technology does not disturb the process nor destroy the process material. If the flow meter is connected to the monitoring interface, the measured number (flow rate) is continuously recorded and monitored in real time. Because the flow meter is installed in the flow of the off-gas, without disrupting the flow, the installation will not cause a problem. With respect to TRL, because the flow meter is a commercialized product for general use in various fields, it can be applied to this process upon completion of testing under pyroprocessing operating conditions. The test should include durability and life expectancy of the equipment. Though the flow meter is not extremely delicate, it may be necessary to change the equipment material or design to increase its life expectance and retain its performance in a severe environment. Because the unit of measurement is 'rate', some criteria are not related to this technology.

To ensure that the off-gas flows to the pipeline connected to the outside of the system, a gas detector can be used. When diagnosing the oxide reduction process, it may be helpful to combine the operation of a flow rate detector with a gas concentration detector. The extent the reaction can be estimated by verifying that pure oxygen gas is discharged outside of the system at a certain rate. The process or process material is not influenced because the gas sensor is installed in the flow of the gas. The measured concentration can be read in real time by connecting the gas sensor to the monitoring interface. Though the limit of detection depends on the performance of the device, the 'ppm' level which is generally used with these devices will be enough for this analysis. When considering that micro-size sensors exist, the required area for installation is minor, the sensor can be located in the middle of the pipeline, and the installation is relatively uncomplicated. Gas detectors are already in commercial use. In addition, detectors developed for nuclear energy facilities, are resistant to the radiation environment and are readily available. Therefore, this technology is expected to have sufficiently high TRL and equipment durability. As identified in the flow meter discussion, some criteria do not apply to this technology.

In the evaluation for other candidate technologies, the selectivity, sensitivity, and accuracy criteria were considered only when the technology is related to a qualitative analysis (Concentration and composition of salt). In this case, because the allocated number was not high (unsuitable or inaccurate), the total score is relatively low contrastive to the other candidates.

4. Conclusion

The current study proposed a set of assessment criteria for the evaluation of PM technologies to pyroprocessing safeguards. The criteria were applied to the process of oxide reduction as an example. The current list may need to be expanded in the future work. For example, DA analysis technology is typically used in NMA rather than PM technology where real time detection is important. To evaluate these technologies, it is necessary to compile a more detailed group of assessment criteria which are specifically related to performance and function.

Additional research is necessary to validate the

Table I. Adaptation assessment criteria to the oxide reduction process

	Process /process material interference or destruction	Possibility of real time detection	Detection Limit	Degree of precision and sensitivity	Possibility of selective analysis	Ease of installation	Generation of secondary waste	TRL	Durability and life expectancy of equipment	Total
Flow rate	1	1				1	1	1	0.5	0.917
Gas concentration	1	1	1			1	1	1	1	1.000
Power supply	1	1				1	1	1	1	1.000
Temperature	1	1				1	1	1	1	1.000
Rotation speed	1	1				0.5	1	1	1	0.917
Potential, current, or voltage	1	1				1	1	1	1	1.000
Concentration And composition of salt	1	0	0.5	0.5	1	0	0	1	0.5	0.500

criteria according to the needs of each unit process, perhaps based on expert elicitation and/or international collaboration with other expert organization(s). These advanced assessment criteria will serve a useful guideline for selecting appropriate candidate PM technologies for pyroprocessing safeguards. Based on the results of using these evaluation criteria, the optimum technologies can be successfully selected for use at a large scale pyroprocessing facility.

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REFERENCES

[1] D. Rappleye, M., Simpson, R, Cumberland, D. McNelis, and M.S. Yim, Simulated real-time process monitoring of a molten salt electrorefiner. Nuclear Engineering and Design, Vol. 273, pp. 75-84, 2014
[2] IAEA Safeguards Glossary, 2001.
[3] S.M. Jeong, B. H. Park, J. M. Hur, C. S. Seo, H. S. Lee, and K. C. Song, An experimental study on an electrochemical reduction of an oxide mixture in the advanced spent-fuel conditioning process, Nuclear Engineering and Technology, Vol. 42(2), pp. 183-192, 2010.
[4] DoD, U. S., Technology readiness assessment (TRA) guidance, Revision posted, 13, 2011.
[5] C. Baker, T. M. Pfeiffer, S. D. Hermann, S. B. Park and S.-H. Kim, Development of a kg-Scale Oxide Reduction Module for Spent Light Water Reactor Fuel, in 2012 International Pyroprocessing Research Conference, Fontana, 2012.