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Gamma-ray Spectra Analyses of Molten Salts in Spent Nuclear Fuels Pyroprocessing Facilities for Mass Measurement

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Abstract

To evaluate radioactive tracer dilution and gamma spectroscopy as a safeguards technique for monitoring the mass of liquid salt in molten salt systems, the gamma spectroscopy data of salt samples from the pyroprocessing facilities in Idaho National Laboratory were acquired and analyzed. The primary focus is the uncertainty analysis of the gamma radioactivity of ^{154}Eu isotope in the molten salts. The facilities include an electrorefiner for pyroprocessing spent oxide fuels (SOF-ER) and Mark-IV ER for processing the metallic fuels from Experimental Breeder Reactor-II (EBR-II). The relative gamma radioactivity uncertainty was consistently at 3% for ^{137}Cs for both Mark-IV ER and SOF-ER salts. However, for ^{154}Eu, it was 8% for SOF-ER salt and 7% for Mark-IV ER salt. The main reason for the higher uncertainty for ^{154}Eu is believed to be the lower counting statistics in gamma spectroscopy. To improve the gamma radioactivity uncertainty of isotope ^{154}Eu, a longer data acquisition time (12 hours instead of 4 hours) and a higher count rate gamma detector for gamma spectroscopy were tested and it was found that the uncertainty for ^{154}Eu was significantly improved from 7% or 8% to 3%.

Keywords: Safeguards, Radioactive Tracer Dilution, Gamma-ray Spectroscopy, Uncertainty, Na-22, Molten Salts

Introduction

Molten salts, in which fissile nuclear materials (NM) accumulate over time, are utilized in the pyroprocessing of spent nuclear fuels (*e.g.*, the electrochemical reduction of spent uranium oxide nuclear fuel and uranium electrorefining) [1–3]. Other broader applications of molten salt are found in the nuclear energy field where it was used as a liquid fuel salt or coolant salt in a molten salt reactor (MSR) [4, 5], or as solar energy storage medium for non-nuclear applications [6]. NM-containing molten salts fundamentally necessitates a different approach to nuclear material accountancy and/or safeguards than do traditional solid fuels. Given the reality that the NM-containing molten salt used in both pyroprocessing and MSRs will necessarily stay in the liquid phase, measurement of the nuclear material inventory for the purpose of safeguards is very challenging. The quantity of the molten salt mass must be known to determine the NM inventory. In general, the mass can be determined by measuring the volume and density of the molten salts. But frequently, the irregular or complicated shapes of molten salt containers make accurate volume measurements very difficult in pyroprocessing systems. This is particularly true for MSRs, in which the salt continuously flows through pipes and heat exchangers, and the temperature differential between the reactor core and heat exchangers may affect the density significantly [7]. It would also be very difficult —perhaps even erroneous—to assign a single density to the salt in an MSR due to the variability caused by fission and off-gassing, and in molten salt spent nuclear fuels pyroprocessing facilities due to the fission products decay buildup.

In recent years, an innovative technique called radioactive tracer dilution (RTD) coupled with gamma spectroscopy, was proposed in Idaho National Laboratory (INL) and evaluated in collaboration with researchers at The Ohio State University as a potential technique for addressing the challenges associated with molten salt mass measurement in molten salt systems [8–10]. In the study, by adding a small amount of ^{22}NaCl into molten LiCl-KCl salt and then conducting gamma spectrum analysis of ^{22}Na in salt samples by a High Purity Germanium (HPGe) detector, the radioactivity of ^{22}Na isotope was found to be linearly proportional to the mass of the salt sample, with an uncertainty level of about 5%. This suggests that the proof of concept of RTD was demonstrated and it may be a promising technique for molten salt mass measurement. The fission products ^{137}Cs and ^{154}Eu were also found to significantly affect the accuracy in measuring the 1274.54 keV gamma-ray emitted by ^{22}Na , due to a direct overlap of one of the gamma-ray peaks from ^{154}Eu (1274.43 keV) and the high Compton plateau and instrument deadtime caused by ^{137}Cs. A diluted salt sample with ^{22}NaCl tracer addition may alleviate these interferences. It is unknown how the gamma radioactivity of a diluted salt sample would respond to the concentration of ^{22}NaCl, although the radioactivity is expected to be linearly proportional to the ^{22}NaCl concentration.

It was proposed that RTD coupled with gamma spectroscopy may be a feasible approach to measure the salt mass in the molten salt vessels for pyroprocessing or in reactor vessels for MSRs. Because the accuracy of salt mass measurements involving RTD is dependent on the radioisotope’s gamma ray radioactivity measurement by gamma spectroscopy, it would be helpful to review and evaluate the existing gamma spectra data of salt samples taken from different molten salt systems, as well as how to address the limitations, if any, associated with gamma spectroscopy. Thus, we analyzed the gamma spectra data for the salt samples taken from the electrorefiner for the pyroprocessing of spent oxide fuels (SOF-ER) and Mark-IV ER vessels in the past years. SOF-ER was used to refine the uranium that was reduced from uranium oxide fuels. In the reduction of oxide fuels, the uranium oxide is electrochemically reduced to uranium metal in a LiCl-1wt% Li_2O electrolyte at around 650°C. Then the uranium metal is subjected to a vacuum distillation process to remove the LiCl salt sticking to the surface of uranium metal. Mark-IV ER was used to treat the used metallic fuels such as U-10Zr used in the Experimental Breeder Reactor-II (EBR-II) [2, 11]. The electrolyte used in SOF-ER and Mark-IV ER is LiCl-KCl-UCl_3 salt. Details about the pyroprocessing of spent oxide fuels and EBR-II fuels can be found elsewhere [1, 11, 12]. Because the fuels processed by the two ERs differ significantly, different levels of fission products accumulated in the vessels during the operations of the two ERs over the years; therefore, these salts could be considered good candidates for understanding the effects of fission products on the molten salt mass measurement by RTD and gamma spectroscopy. One important fission product is ^{154}Eu which has a number of gamma-ray peaks in the gamma spectrum. One of its peaks at 1274.43 keV coincides with the only gamma-ray emission from ^{22}Na at 1274.54 keV that is used to determine the radioactivity of ^{22}Na. The uncertainty of ^{154}Eu radioactivity measurements by gamma spectroscopy is expected to directly affect the accuracy of ^{22}Na radioactivity, and thus the accuracy of the salt mass measured by RTD.

The primary objectives of the present study are to analyze the existing gamma spectroscopy data of the salt samples from SOF-ER and Mark-IV ER and to evaluate the feasibility and measurement uncertainty of RTD based on gamma spectroscopy for safeguards purposes. During the data review process, it was found that the uncertainty for ^{154}Eu isotope, the isotope of high interest, was quite high for safeguards purposes (to be provided in Results and Discussion section), some approaches were proposed and tested to improve the uncertainty of ^{154}Eu. The ultimate goal is to develop a molten salt mass measurement technique that is based on RTD and gamma spectroscopy for nuclear material accountancy in high temperature molten salts such as those used in MSRs and pyroprocessing facilities for recycling spent nuclear fuels.

Principle of Molten Salt Mass Measurement by RTD and Gamma Spectroscopy

The molten salt mass measurement by RTD and gamma spectroscopy for a molten salt system (homogeneous liquid salt) is based on the measurement of radioactivity of isotopes including radioactive tracer added into the molten salts by gamma spectroscopy. In gamma spectroscopy systems, the gamma rays emitted from a sample are detected by using solid state semiconductor detectors such as the HPGe detector. Well-resolved characteristic peaks can be used for quantitative analysis of isotopes. Data acquisition and analysis of isotopes can be conducted using commercial softwares such as Genie 2000 from Mirion Technologies. Details about gamma spectroscopy can be found elsewhere [13], [14].

Using the tracer ^{22}Na (in ^{22}NaCl salt form)—a radioisotope with strong gamma emission and a half-life of 2.6 years—in this study as an example, the molten salt mass measurement for a molten salt system by RTD and gamma spectroscopy can typically be achieved by the following process:

(1) Tracer salt (or ^{22}Na-tracer bearing salt) preparation: Tracer salt that contains ^{22}NaCl and carrier salt (such as LiCl-KCl eutectic salt used in this study) is made by mixing and melting ^{22}NaCl and LiCl-KCl salt, followed by solidification of the melt and breaking the solidified salt ingot into small pieces.

(2) Tracer salt sampling: Multiple tracer salt samples are taken for gamma spectroscopy measurement.

(3) Salt sampling before tracer salt addition: Before the tracer salt is added into the molten salt, multiple salt samples are taken from the molten salt vessel.

(4) Tracer salt dilution: A small amount of tracer salt is added into a vessel that contains a large amount of molten salt. Note that the mass of the molten salt in the vessel is unknown.

(5) Salt sampling after tracer salt addition: After the tracer salt is added into the molten salt, multiple salt samples are taken from the molten salt vessel.

(6) Gamma spectroscopy measurement of tracer salt samples and salt samples taken from the molten salt vessel before and after tracer salt addition.

(7) Calculation of the molten salt mass: The mass of the molten salt in the vessel can be determined by the mass of the tracer salt added into the molten salt, the gamma radioactivity of ^{22}Na tracer per unit mass in the tracer salt, the gamma radioactivity of ^{22}Na tracer per unit mass in the molten salt before and after tracer salt addition. Based on the activity conservation rule, the mass of the molten salt in the vessel can be calculated by

(1)

where is the mass (in gram) of the unknown molten salt in the vessel, is the mass (in gram) of the tracer salt added into the molten salt, is the gamma radioactivity from ^{22}Na isotope per unit mass (in MBq/g) for the molten salt in the vessel before tracer salt addition, and are the gamma radioactivity from ^{22}Na isotope per unit mass (in MBq/g) for the tracer salt and for salt samples taken from the vessel after tracer salt addition, respectively. Therefore, the mass (M) of the molten salt in the vessel is . Because the tracer salt mass (m) is negligible as compared with the molten salt mass (M) in the vessel, the activity per unit gram (A) for the tracer salt is significantly greater than that for the salt sample, that is A>>a, as an approximation,

(2)

Materials and Methods

For the gamma spectroscopy, standard procedures with good quality assurance were applied to all the salt samples collected from the pyroprocessing facilities over the past two years. Typically, the salt samples, usually in 1 to 2 grams for each sample, were collected from SOF-ER and Mark-IV ER, and then transferred to the hot cell of Analytical Laboratory of INL for gamma spectrum counting. To prepare the salt samples for gamma spectroscopy, a small amount of salt was weighed and then dissolved in water of known volume to make diluted salt solutions. Often, a very high dilution factor is required. This is to reduce the dose rate so that the samples are not only able to be handled safely by lab personnel, but also to ensure that the sample emission rate will yield quality data. Too high a count rate increases the dead time of the detector, leading to sum peaks and poorly shaped peaks. With too low count rate, important species may be below detection limits. For gamma-spectroscopy, the ^{137}Cs peak is the dominant peak in the spectrum and will be the driving factor between what is “too low” and “too high” in count rate. Diluted salt solutions of 1-ml volume were used for gamma spectroscopy, and 1-ml standard solution was used to calibrate the spectrometer. The standard data acquisition time was 4 hours. The uncertainty of the gamma radioactivity of isotopes was determined by the two-sigma (2σ) method. Figure 1 shows the picture of the gamma spectrometer used in the study.

Figure 1 Photograph of the gamma spectrometer outside the hot cell in Analytical Laboratory

Following the uncertainty analysis of the gamma spectra for salt samples from Mark-IV ER and SOF-ER, it was found that the overall uncertainty @ 2σ for ^{154}Eu is quite high, likely due to low counts in its gamma peaks. To improve the statistics in subsequent measurements, the acquisition time was increased from 4 hours to 12 hours, and a high efficiency detector was used. The improvement of the uncertainty of the gamma spectra measurement was verified using two existing salt samples collected from Mark-IV ER.

Results and Discussion

Gamma spectroscopy of molten salts from spent nuclear fuel pyroprocessing facilities of INL

Salt samples from Mark-IV ER and SOF-ER were periodically sent to Analytical Laboratory for isotopic analysis. Generally, inductively coupled plasma mass spectrometry (ICP-MS) was performed for all salt samples, while gamma spectroscopy was performed for some select samples. In gamma spectroscopy, the isotopes including ^{241}Am, ^{144}Ce, ^{60}Co, ^{134}Cs, ^{137}Cs, ^{154}Eu, ^{155}Eu, ^{54}Mn, ^{106} Ru/Rh, and ^{125}Sb were analyzed. Table 1 and Table 2 list the results of a salt sample from Mark-IV ER and SOF-ER, respectively. Note that ^{22}Na radioisotope was not analyzed because the radioactivity from ^{22}Na was zero or negligible.

Table 1 Isotopic analysis of a driver salt sample from Mark-IV ER by gamma spectra analysis

Analyte	Radioactivity (MBq/g)	Relative Uncertainty @ 2σ
^{241}Am	<11.1	N/A
^{144}Ce/Pr	<37	N/A
^{60}Co	<0.74	N/A
^{134}Cs	<1.1	N/A
^{137}Cs	8.9×10^3	±3%
^{154}Eu	10.6	±10%
^{155}Eu	<7.4	N/A
^{54}Mn	<1.1	N/A
^{106}Ru/^{106}Rh	<29.6	N/A
^{125}Sb	<18.5	N/A

Table 2 Isotopic analysis of a salt sample from SOF-ER by gamma spectra analysis

Analyte	Radioactivity (MBq/g)	Relative Uncertainty @ 2σ
^{241}Am	114.3	±3%
^{144}Ce/Pr	<0.4	N/A
^{60}Co	<0.07	N/A
^{134}Cs	<0.07	N/A
^{137}Cs	16.8	±3%
^{154}Eu	0.3	±9%
^{155}Eu	<0.1	N/A
^{54}Mn	<0.07	N/A
^{239}Np	<0.8	±5%
^{243}Am	<0.7	±5%

It can be seen from the analysis that the isotopes in Mark-IV ER and SOF-ER salts are significantly different. This is expected because the two ERs process two different types of spent fuels. In the Mark-IV ER salt, the gamma radioactivity per gram is mainly from ^{137}Cs, which is 8.9×10^3 MBq/g. ^{154}Eu has a radioactivity of 10.6 MBq/g, with an uncertainty level of 10%. In SOF-ER salt, the gamma radioactivity per gram is mainly from isotopes ^{241}Am (114.3 MBq/g) and ^{137}Cs (16.8 MBq/g), while the gamma radioactivity from ^{154}Eu is much lower (0.3 MBq/g), with an uncertainty level of 9%.

Because the gamma radioactivity of ^{154}Eu and other isotopes with high gamma activities in the salts is thought to potentially be the main factor that may affect the accuracy and uncertainty of RTD for molten salt mass determination, only ^{241}Am, ^{137}Cs, and ^{154}Eu in the SOF-ER salt and ^{137}Cs and ^{154}Eu in the Mark-IV ER salt were selected for further review and discussion.

Table 3 and Table 4 represent the gamma spectra data for different salt batches, collected over the past two years, from SOF-ER and Mark-IV ER, respectively. As can be seen, in SOF-ER salts, the total gamma radioactivity per gram is in the range of 56.2 MBq/g to 131.6 MBq/g. The uncertainty for most ^{241}Am and ^{137}Cs radioactivity measurement is 3%. The SOF-ER salt contains a small amount of ^{154}Eu with a radioactivity range of 0.2–0.4 MBq/g, but the uncertainty for ^{154}Eu scattered significantly, in the range of 4 –14%, with an average uncertainty of 8%. Because the 1274.43 keV gamma peak from ^{154}Eu has the same energy as the only peak from ^{22}Na tracer, the presence of ^{154}Eu and its high uncertainty are expected to affect the accuracy of salt mass measurement by RTD and gamma spectroscopy. Thus, improvement of uncertainty for ^{154}Eu radioactivity is needed to effectively eliminate its contribution to the ^{22}Na gamma peak (1274.54keV) counts and thus enable accurate determination of the ^{22}Na tracer radioactivity.

In the Mark-IV salt samples, the gamma radioactivity per gram, in the range of 8.2–8.9 $\times 10^3$ Mbq/g, is much higher than for the SOF-ER salt. The uncertainty for ^{137}Cs radioactivity is as low as 3%. The Mark-IV ER salt samples feature ^{154}Eu radioactivity of 6.6-10.6 MBq/g. As with the high level and wide range of the uncertainty for ^{154}Eu radioactivity in the SOF-ER salt, the uncertainty for ^{154}Eu in the Mark-IV ER salt samples also showed significant variation, in the range of 6–10%, with an average of 7%. Again, the uncertainty for the ^{154}Eu isotope needs to be improved.

Table 3 Gamma spectra data for SOF-ER salt samples

Sample #	^{241}Am radioactivity (MBq/g) and uncertainty @ 2σ	^{137}Cs radioactivity (MBq/g) and uncertainty @ 2σ	^{154}Eu radioactivity (MBq/g) and uncertainty @ 2σ
SOF-ER 1	42.6, ±3%	14.7 , ±3%	0.4 ±7%
SOF-ER 2	40.3, ±3%	15.5 , ±3%	0.4, ±4%
SOF-ER 3	41.1, ±3%	15.3, ±5%	0.4, ±7%
SOF-ER 4	114.0, ±3%	17.4 , ±3%	0.2, ±14%
SOF-ER 5	114.3×10^3, ±3%	16.8, ±3%	0.3, ±7%

Table 4 Gamma spectra data for Mark-IV ER salt samples

Sample #	^{137}Cs radioactivity (MBq/g) and uncertainty @ 2σ	^{154}Eu radioactivity (MBq/g) and uncertainty @ 2σ
Mark-IV ER1	8.8×10^3, ±3%	7.0, ±7 %
Mark-IV ER2	8.6×10^3, ±3%	6.6, ±6 %
Mark-IV ER3	8.2×10^3, ±3%	6.7, ±6 %
Mark-IV ER4	8.4×10^3, ±3%	7.0, ±6 %
Mark-IV ER5	8.9×10^3, ±3%	10.6, ±10 %
Mark-IV ER6	8.7×10^3, ±3%	8.2, ±8 %
Mark-IV ER7	8.2×10^3, ±3%	8.4, ±6 %

Improvement of the uncertainty in gamma spectroscopy

After analyzing the gamma spectra data, the high uncertainties for ^{154}Eu were believed to mainly stem from the relatively small number of counts. Even after 4 hours of counting in gamma spectroscopy, the peaks were found to be a few hundred counts. As the counting uncertainty is proportionate to the square root of the number of counts, so small peaks can have a large effect on the overall uncertainty. By increasing the data acquisition time and using a detector that has a higher efficiency, counts for all peaks can be increased, potentially improving the measurement uncertainties. Due to the large amount of ^{137}Cs in the sample, counting a more concentrated sample is most likely not a feasible alternative, and would likely result in a degradation of data quality. A higher concentration of ^{137}Cs would result in an increased amount of Compton scattering from the ^{137}Cs gamma rays, obscuring most of the spectrum below 662 keV. Additionally, the dead time would rise, resulting in sum peaks, and possibly distorted peak shapes.

To improve the uncertainty for ^{154}Eu radioactivity, we increased the data acquisition time from 4 hours to 12 hours and used a detector with higher efficiency, thus enabling the accumulation of a sufficient number of counts for all ^{154}Eu peaks. As an example, Figure 2 shows the count rate for a Mark-IV salt sample that was analyzed using a detector with higher efficiency during gamma spectra measurements for 4 hours and 12 hours. Table 5 lists the counts for each peak obtained from the two different gamma spectrum measurements. As can be seen, the counts at 1274.43 keV in 12-hr test are ~16 times those in the 4-hr test.

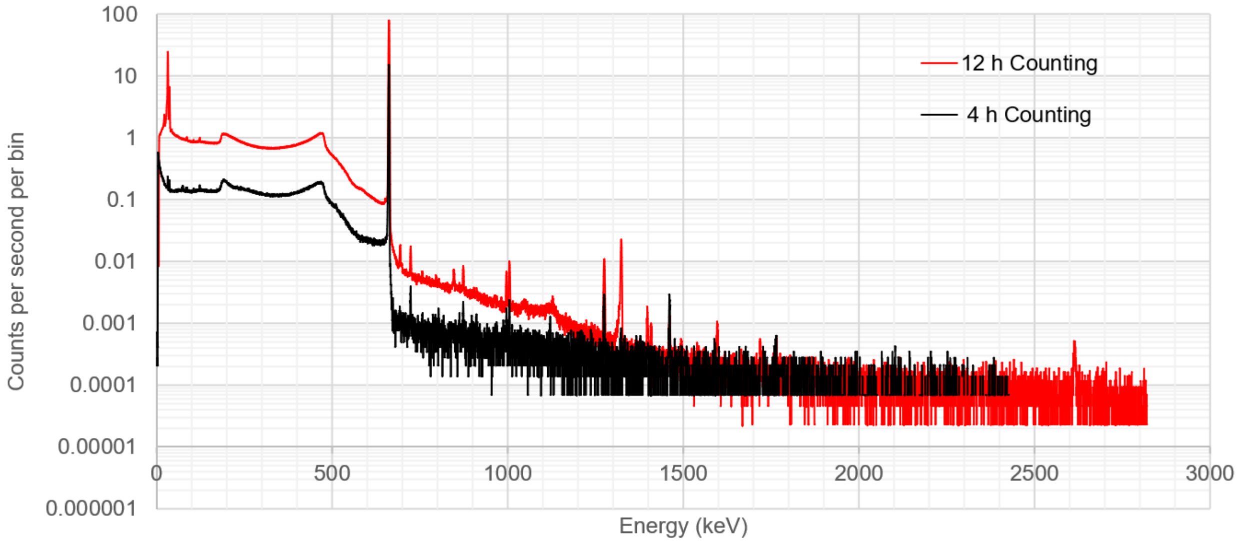

Figure 2 Comparison of the counts rates obtained using two different detectors during the gamma spectrum measurements of a Mark-IV ER sample with data acquisition times of 4 and 12 hours, respectively.

Table 5 Counts per peak in gamma spectra measurements for 4 hrs and 12 hrs

Gamma-ray energy keV	Counts per peak in 4-hr test	Counts per peak in 12-hr test
123	1740 ± 248.1	27700 ± 1289.3
723	231 ± 33.8	3420 ± 157.3
873	125 ± 27.1	1800 ± 123.3
1004	174 ± 27.2	2900 ± 124.5
1274	295 ± 32.1	4700 ± 134.8

Table 6 shows the gamma spectra data for two Mark-IV salt samples that were tested using 12 hrs data acquisition. As expected, the uncertainty for gamma spectra measurement has been significantly improved from 6% and 7% for different peaks (as seen in Table 5) to consistently for all at 3%. An uncertainty of 3% is the best measurement limit achievable for the gamma spectrometer for radioactive salt measurement used in this study.

Table 6 Gamma spectra for two Mark-IV ER salt samples with data acquisition of 12 hours

Sample #	^{137}Cs radioactivity (MBq/g) and uncertainty @ 2σ	^{154}Eu radioactivity (MBq/g) and uncertainty @ 2σ
Mark-IV ER1	9.3×10^{3} , ±3%	7.2 MBq/g, ±3 %
Mark-IV ER3	8.1×10^{3} , ±3%	7.0 MBq/g, ±3 %

Analysis of uncertainty of gamma spectroscopy and its effects on its potential application for molten salt mass determination by radioactive tracer dilution

Based on the principle of molten salt mass measurement by RTD coupled with gamma spectroscopy, the salt mass of a molten salt system is calculated based on the mass of the tracer salt added and the gamma radioactivity of ^{22}Na tracer per gram for the tracer salt and for the salt samples taken from the molten salt system before and after tracer salt addition. Therefore, the uncertainty of the salt mass measurement is affected by the gamma spectroscopy uncertainty (for both tracer salt and salt samples) as well as the uncertainty of the tracer salt mass measurement. Uncertainty propagation exists in this situation. When uncertainty is considered, Equation 2 for salt mass determination by RTD can be written as

(3)

where and are respectively the absolute uncertainty for the salt mass measured via RTD coupled with gamma spectroscopy and the mass of the tracer salt added. and are respectively the absolute uncertainty for gamma radioactivity per gram of ^{22}Na isotope in the tracer salt added, in the salt sample taken from the molten salt vessel before tracer salt addition, and in the salt sample from the vessel after tracer salt addition. During gamma spectroscopy analysis of the salt samples, the ^{22}Na from the tracer salt has only one peak at 1274.54 keV which is used for the gamma radioactivity for tracer salt. But the ^{154}Eu isotope in the salt samples also has a peak at 1274.43 keV. Assuming the uncertainty for the ^{22}Na tracer is the same as for ^{154}Eu, and because, in this study, the ^{22}Na radioactivity (a_0) in the molten salt vessel before tracer salt addition is zero (or negligible), the overall relative uncertainty for the salt mass measurement can be expressed as

(4)

The absolute uncertainty for the tracer salt mass measurements in a hot cell is usually less than 0.01g. For 1g or more of tracer salt added, the relative uncertainty is 1% or lower, which is significantly lower than the uncertainty for the gamma spectroscopy. An uncertainty of 1% was used here for calculations. For the gamma spectroscopy involving data acquisition time of 4 hours, an average relative uncertainty of 7 % for ^{22}Na was considered. The relative uncertainty ()

for the salt mass measurement would be =10%, which is quite high for accurate salt mass measurement. For the gamma spectroscopy involving data acquisition time of 12 hours and a higher count rate detector, the relative uncertainty 3% for ^{22}Na was used, and the relative uncertainty () for the salt mass measurement would be = 4%. Thus, the relative uncertainty for the salt mass measurement was significantly improved.

It should be noted that the calculated uncertainty for the gamma spectroscopy itself can be lower than 3%. But because the standard used for calibrating the gamma spectrometer carries an uncertainty level of 3%, the uncertainty for the gamma spectroscopy was always reported as being at least 3%. Therefore, the above overall 4% uncertainty for the salt mass measurement by RTD is currently the lowest value achievable by our system.

To further improve the accuracy of salt mass measurement via RTD and gamma spectroscopy, a new standard with lower uncertainty level is needed for calibrating gamma spectrometer. For the sake of discussion, if a new calibration standard with an uncertainty level of 1% can be procured and used, and the calculated uncertainty for ^{22}Na from gamma spectroscopy is 1.5% — as is potentially achievable based on current best gamma spectroscopy practices—the uncertainty level for the salt mass measurement calculated from Equation 4 would be as low as 2%.

The uncertainty in salt mass measurement by RTD coupled with gamma spectroscopy is an important research area for developing it as a safeguards technology for molten salt systems. In addition to the improvement of uncertainty by increasing data acquisition time, the largest improvement of salt mass uncertainty may be achieved by a significant increase of ^{22}Na tracer radioactivity to make the interfering peak from ^{154}Eu insignificant, which is being investigated. Another derivative method is to increase the volume of the salt solution or sample size, while duly accounting for the self-shielding effect. This would increase the counts for ^{22}Na gamma peak, thus improving the uncertainty level.

Conclusion

A molten salt mass measurement technique based on radioactive tracer dilution (RTD) coupled with gamma spectroscopy was investigated for safeguards purposes—both in terms of its feasibility and uncertainty for salt mass measurement using ^{22}Na tracer— by analyzing the existing gamma spectroscopy data of salt samples from INL spent fuel pyroprocessing facilities. The gamma spectra data for salt samples from Mark-IV ER and SOF-ER—used for pyroprocessing EBR-II metallic fuels and spent oxide fuels respectively—were analyzed. The gamma radioactivity of SOF-ER salts is much lower than that of Mark-IV ER salts. The gamma radioactivity in SOF-ER salt mainly stems from ^{241}Am and ^{137}Cs, while the radioactivity of the ^{154}Eu is quite low: 0.2–0.4 MBq/g. The high gamma radioactivity of the Mark-IV ER salt predominantly stems from ^{137}Cs. The radioactivity of ^{154}Eu in Mark-IV ER salt is 6.6–10.6 MBq/g. The uncertainly for the ^{154}Eu is generally 6–10% for Mark-IV ER salt and 4–14% for SOF-ER salt.

We identified the factors affecting the uncertainty of molten salt mass determination by RTD coupled with gamma spectroscopy. To improve the uncertainty in gamma radioactivity measurement of ^{154}Eu isotope, options such as extended data acquisition time and higher detector count rates in gamma spectroscopy, higher tracer radioactivity, and large sample sizes were identified. In the current analysis, by increasing the data acquisition time from the standard 4 hours to 12 hours and using a detector with a higher count rate, the uncertainty @ 2σ for the ^{154}Eu isotope in two Mark-IV ER salt samples was significantly improved from 6% and 7% to 3%. For developing RTD coupled with gamma spectroscopy as an accurate, reliable safeguards technology for monitoring salt mass in molten salt systems, further improvement of uncertainty is needed, and this remains a subject currently under investigation.

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