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MATRIX CHARACTERIZATION OF PLUTONIUM RESIDUES BY ALPHA-PARTICLE SELF-INTERROGATION

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Abstract
Legacy plutonium residues often have inadequate item descriptions. Nondestructive characterization can help segregate these items for reprocessing or provide information needed for disposal or storage. Alpha particle-induced gamma-ray spectra contain a wealth of information that can be used for matrix characterization. We demonstrate how this information can be used for item identification. Gamma-ray spectra were recorded at the Los Alamos Plutonium Facility from a variety of legacy, plutonium-processing residues and product materials. The comparison and analysis of these spectra are presented.

INTRODUCTION
Knowledge of the composition of feed material may be required for processes used in the recovery, stabilization, and disposal of plutonium in residues. For example, some materials, including fluorides and volatile compounds, are excluded from processes envisioned for the disposition of plutonium by immobilization. Limits on the concentration of feed impurities for these processes are currently being developed. Because some portion of plutonium items have incomplete descriptions, identification of impurities using item descriptions is not always possible. Consequently, a nondestructive diagnostic tool is desired for matrix characterization.

Information needed to characterize matrix material can be obtained using gamma-ray signatures from sample self-interrogation and neutron activation analysis. The information contained in these signatures is incomplete in the sense that it does not provide the chemical form of matrix constituents. However, elemental analysis is often sufficient to categorize residues. From self-induced gamma-ray signatures, isotopes of various light elements can be identified, including beryllium, boron, oxygen, fluorine, sodium, magnesium, aluminum, silicon, chlorine, and potassium. From neutron activation analysis, additional elements such as hydrogen and iron can be identified. By detecting these elements and determining their relative abundance, residues can be identified and categorized. Some materials of interest such as fluorides can be identified directly. The presence of other materials such as NaCl can be inferred by the detection of its constituents and available process knowledge.

Gamma-ray signatures from self-interrogation provide the most complete information on the composition of plutonium residues. Radioactive isotopes found in plutonium residues are prolific alpha emitters. When these isotopes are intimately mixed with the matrix, alpha particles (often in excess of 5 MeV) induce the production of gamma rays in the matrix material. High-energy gamma rays, typically greater than 1 MeV, are produced by (α,n), and (α,p) reactions with light elements and also by Coulomb excitation. Because the number of decayed gamma rays above 1 MeV is relatively small, the sensitivity of self-interrogation is high even in the presence of multiple
Fig. 1. Passive gamma-ray spectra for plutonium bearing materials from 300 keV to 3 MeV. A background spectrum is shown for comparison. The line labeled $^{208}\text{TI}$ corresponds to a decay chain ending in $^{208}\text{Pb}$.

Fig. 2. Passive gamma-ray spectra for electrorefining salts from 400 keV to 3.6 MeV.
that there are differences in the matrices. It is interesting that those salts without sodium peaks have a prominent, broadened peak at 478 keV. This peak could correspond to boron through the $^{10}\text{B}(n,\alpha)$ reaction or to lithium through the $^7\text{Li}(\alpha,\alpha')$ reaction. Improved active neutron interrogation could resolve this ambiguity.

CONCLUSIONS AND FUTURE WORK
We have demonstrated the ability to differentiate plutonium residue matrices by self-interrogation signatures. Alpha-particle-induced gamma rays enable the detection of a variety of light elements. In this preliminary study, we observed lithium, beryllium, boron, oxygen, fluorine, sodium, magnesium, aluminum, chlorine, and potassium. However, it is clear from earlier studies[1] that many other elements can be detected, including silicon, phosphorous, manganese, tungsten, platinum, tantalum, and vanadium, some of which may be useful for characterizing plutonium residues. Improvement of the active interrogation mode, by increasing neutron source intensity or by improving counter design, will enable complementary measurements. Active interrogation is necessary, for example, to detect hydrogen which is inaccessible by self-interrogation and other elements that have low sensitivity.

Quantitative analysis of elemental concentration may be possible. For example, if the abundance of alpha-emitting isotopes is known (e.g., through gamma-ray isotopic analysis) and assumptions can be made about the chemical form of the material, then bounds on the concentration of elements for which alpha-particle-induced gamma rays are observed can be estimated. The accuracy of the analysis will depend on the quality of thick target yields available in the literature. The application of quantitative analysis for screening purposes as well as a more detailed investigation of the sensitivity of self-interrogation signatures is underway.

REFERENCES

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