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Background X-ray Spectrum of Radioactive Samples

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INTRODUCTION

An energy-dispersive X-ray spectrometer (EDS) is commonly used with a scanning electron microscope (SEM) to analyze the elemental compositions and microstructures of a variety of samples. For example, the microstructures of nuclear fuels are commonly investigated with this technique. However, the radioactivity of some materials introduces additional X-rays that contribute to the EDS background spectrum. These X-rays are generally not accounted for in spectral analysis software, and can cause misleading results. X-rays from internal conversion [1], Bremsstrahlung [2] radiation associated with alpha ionizations and beta particle interactions [3], and gamma rays from radioactive decay can all elevate the background of radioactive materials.

Internal conversion of short lived alpha emitters causes X-rays whose energies are characteristic of the daughter isotopes, increasing the intensities of these X-rays and possibly causing their concentrations to be over-reported. Bremsstrahlung produces a continuous X-ray spectrum which decreases the signal to noise ratio, thus making identification of statistically significant X-ray peaks difficult [3]. Although the energies of gamma rays are typically too high to be included in the spectrum directly, both gamma rays and X-rays from internal conversion can contribute to the spectrum via secondary interactions such as fluorescence [1, 4].

This research represents an attempt to model the consequences of radioactive processes and use the model to analyze the spectrum from a specific sample.

DESCRIPTION OF WORK

The overall approach taken by this work was to create a model of the expected spectrum from radioactive contributions and then compare this model to experimental results obtained from a real sample. First, a model of the real sample was developed based on the assumption that the sample was homogeneous (i.e. all isotopes were uniformly distributed throughout the sample) and the angular dependence of X-rays was not significant. This model predicted the radiation contribution by alpha particles, beta particles, gamma rays, and internal conversion X-rays. Next, the secondary interactions, namely Bremsstrahlung and fluorescence, were modeled using NISTMonte. Spectral contributions by alpha particle ionization were not modeled since alpha particle transport is dependent on the microstructure, and therefore would not be consistent with the assumption of sample homogeneity. NISTMonte was also used to evaluate the fluorescence contribution from generated X-rays and to model an EDS detector. Finally, a real radioactive sample with composition identical to the modeled composition was placed in an SEM and a beam-off and a beam-on spectrum were recorded using an EDS.

RESULTS

The isotopic composition of this study's sample can be found in Table 1. Since alpha emission is the prominent decay mode for this sample, it was expected that the modeled background would underestimate the experimental spectrum. Regardless, the results of the aforementioned work are presented.

Figure 1 depicts the expected background contributions from various sources compared against the actual beam-on spectrum. Those sources are gamma rays, X-rays (from internal conversion), Bremsstrahlung from beta particles produced by radioactive decay, and Bremsstrahlung produced by the electron beam. Even a small beta source (0.09 wt%) produces copious background X-rays.

The primary discrepancies between the predicted and experimental spectrum are believed to

be attributed to ionizations caused by alpha interactions. The discrepancies exist at lower energy (<12 keV), where alpha ionizations become more frequent as the alpha particles slows down.

From this study it was observed that radioactive samples have X-ray sources that are not accounted for in many spectral analysis programs. This work provides insight into what corrections need to be made to existing spectral analysis programs in order to accurately perform quantitative analysis of radioactive samples using an EDS. Specifically internal conversion X-rays, alpha and beta particle interactions, and gamma ray fluorescence also need to be considered. Depending on the importance of alpha decay, it may also be necessary to model the microstructures in individual samples.

TABLE I. Sample Composition.

Element or Isotope	Weight %	Radiation Contribution
Zr	15.00%	α, γ, X-ray
U^{235}	27.00%	α, γ, X-ray
U^{238}	32.87%	α, γ, X-ray
Np^{237}	1.99%	α, γ, X-ray
Pu^{238}	0.01%	α, γ, X-ray
Pu^{239}	16.50%	α, γ, X-ray
Pu^{240}	3.30%	α, γ, X-ray
Pu^{241}	0.09%	α, β, γ, X-ray
Pu^{242}	0.07%	α, γ, X-ray
Am^{241}	3.00%	α, γ, X-ray

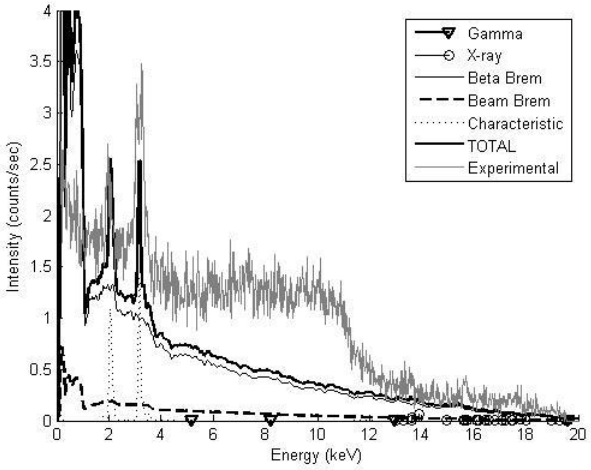

Fig. 1. Comparison of predicted background EDS spectrum and experimental spectrum

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