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MEASUREMENT OF ^{235}U ENRICHMENT USING THE SEMI-PEAK-RATIO TECHNIQUE WITH CdZnTe GAMMA-RAY DETECTOR

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Abstract - In uranium enrichment plants and nuclear fuel fabrication facilities, exact measurement of fissile isotope enrichment of uranium is required for material accounting in international safeguards inspection as well as process quality control. The purpose of this study was to develop a simple measurement system which can portably be used at nuclear fuel fabrication plants especially dealing with low enriched uranium. For this purpose, a small size CZT (CdZnTe) detector was used, and the detector performance in low uranium gamma/X -rays energy range was investigated by use of various enriched uranium oxide samples. New enrichment measurement technique and analysis method for low enriched uranium oxide, so-called, "semi-peak ratio technique" was developed. The newly developed method was considered as an alternative technique for the low enrichment and would be useful to account nuclear material in safeguarding activity at nuclear fuel fabrication facility.

INTRODUCTION

The common methods for isotopic enrichment determination are an infinite sample gamma measurement technique [1,2], and a peak-ratio technique [3,4,5]. The infinite sample technique gives a good accuracy while using a thick uranium sample. This technique needs well designed collimator so that the detector always sees same sample geometry. Normally, the counting rate of 186-keV gamma-ray from ^{235}U is measured. Then, the counting rate is used to determine the enrichment by referencing an enrichment calibration curve determined previously as a function of 185.7-keV ceunting rate[1,2]. The infinite sample technique is not suitable to a low enrichment sample, because the 185.7-keV counting rate is not sufficient to analyze low enrichment samples. To increase the number of counts, such system has to use large volume of detector crystal. As result, the weight of system is heavy, and is not adequate for portable field operation.

To overcome such limitation, the peak-ratio technique has been developed [3,4,5]. This technique needs not any calibration process using enrichment standard and heavy collimator. This technique uses the known branching ratios and the intensities of various uranium isotopes and their decaying daughter isotopes. By fitting and comparison with the known gamma/X-ray nuclear data, the enrichment is determined. In spite that nuclear data is well known, this technique (especially based on CZT detector) showed that the determination error is over 20% for low enrichment samples [3].

Newly proposed technique is an alternative one for determination technique especially for low enrichment samples. This technique suffices two requirements; 1) measuring accuracy, and 2) portability at low enrichment range. The second requirement is important in safeguards

activity, because the facilities under safeguards activities are spread widely. This new technique uses merits of previous two techniques; detection method is similar to that of an infinite sample technique, while analysis method is similar the peak-ratio technique. So it is named as "semi-peak-ratio technique". Only two strong gamma-rays from different uranium isotopes are used to determine isotope enrichment. It needs calibration like infinite sample technique according to uranium enrichment standards, but needs not any collimator system. The determination accuracy is located between those of two techniques. New technique based on CZT detector is an adequate portable system for safeguards field operation.

ENRICHMENT DETECTION PRINCIPLE

Uranium samples to be measured for safeguards inspection is composed of various kinds of isotopes. Generally, natural uranium: consists of ^{238}U (99.27%), ^{235}U (0.720%), and ^{234}U (0.096910). The natural uranium itself can be used as nuclear fuel in HWR (Heavy Water Reactor). In order to use in LWR (Light Water Reactor), however, more fissile isotope, ^{235}U, are needed. So the natural uranium is enriched at enrichment plant typically up to 3 ~ 5 % for LWR use. The ^{235}U isotope is special nuclear material controlled by the international safeguards frame, and therefore an exact 235U mass has to be measured.

The ^{235}U enrichment can be measured if radiation is available and if a few specific measurement conditions are met. The isotopes of uranium emit alpha, beta, neutron and gamma radiation. The primary radiation used in passive nondestructive assay (NDA) of uranium samples is gamma radiation, which is usually dominated by emissions from ^{235}U decay.

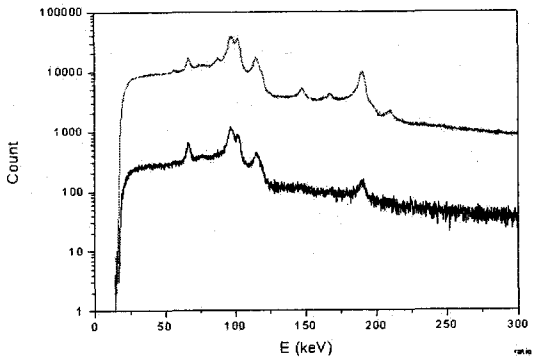

Fig. 1. Uranium sample spectra of ^{235}U enrichments of 3.2% (Upper) and 0.7%(low).

Presently a transportable enrichment system based on the peak-ratio technique was developed, in which some characteristic gamma-rays from uranium isotopes are used for analysis. For examples, ^{238}U decay produces 63.2-keV gamma-ray and ^{235}U decay produces 185.7-keV gamma-ray which is the most prominent single gamma ray from ^{235}U isotope. Fig. 1 is a typical gamma-ray spectra measured by CZT system for UO_2 powder samples with 3.2% and 0.7% enrichment cases, respectively. In the figure 1, the peaks of 185-7-keV and 63.2-keV gamma-rays are populated strongly. It is the signal that the enrichment can be estimated by the peak intensity ratio of 185.7-keV and 63.2-keV.

EXPERIMENTS

The 5 × 5 × 5 cubic mm CdZnTe(CZT) detector, which has moderate energy resolution in room-temperature compared with generally used HP-Ge and NaI detectors. The detector was coupled with 297-CZT probe module(EG&G ORTEC) which consists of charge-sensitive pre-amplifier and amplifier less than 0.5 ?sec shaping time. The output of CZT probe module was connected to an ORTEC MicroNOMAD portable MCA. The MicroNOMAD is a 1.5-lb battery-powered potable gamma-ray spectrometer developed for field operation such as nuclear material safeguard system and environmental

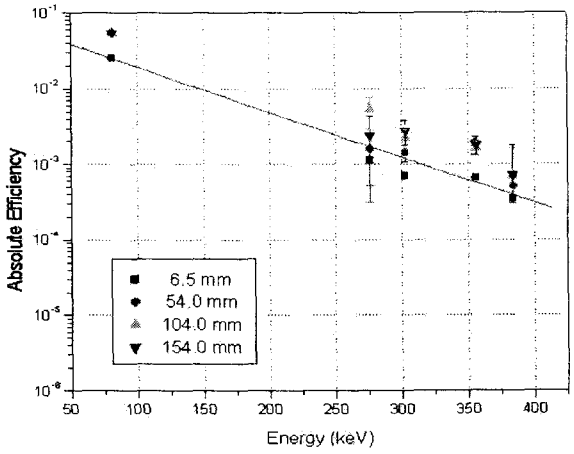

Fig. 2. The absolute detection efficiency of 5x5x5 mm CZT detector. Measurement was performed at different distances. The solid line is eye guide line.

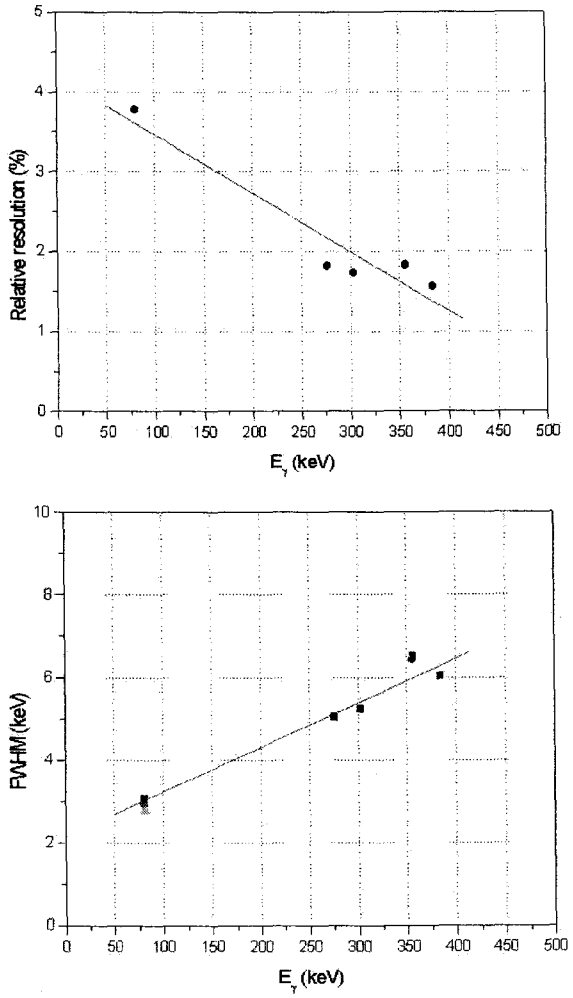

Fig. 3. Measured detection energy resolutions as a function of photo-peak energy (left), and FWHM (right). The solid lines are fitted by linear regression method.

survey. The acquired spectra were downloaded to portable PC with 2k channels and analyzed..

The absolute detector efficiency was measured by using ^{133}Ba gamma-ray standard source that covers gamma-ray energy range from 50 keV to 400keV. The absolute gamma/X-ray detection efficiency is shown in Fig. 2. The determined absolute efficiency is about 1% near 100-keV region especially interested in. To collect geometrical dependence, measurements are performed several times at different distances between source and detector crystal.

CZT detector has better energy resolution than that of NaI(Tl) scintillation detector, so it covered energy resolution range between HP-Ge semiconductor detector and NaI(Tl) scintillation detector. The energy resolution was measured as a function of gamma-ray energy. It is indicated that energy resolution was measured less than 4% of photo-peak energy, and 3-keV FWHM near 100-keV region as shown in Fig.3-(a) and 3-(b), respectively.

DISCUSSION

In the peak-ratio technique, the useful gamma/X-rays categorized into several groups are used. In the present study three gamma/X-ray groups were used. These are 63.2-keV from ^{238}U decay, 185.7-keV from ^{235}U decay, and 98-keV K-shell fluorescence X-rays induced by photoelectric effect of primary gamma-rays in which the intensity of X-ray depends on the bulk mass of uranium. The intensity of 185.7-keV decreases with increase in enrichment. On the contrary the intensity of 63.2-keV increases with decrease in enrichment. The peaks near 98-keV is so complex that it is not adequate peak to analyze because different physical origins are involved to produce it. In fact, it is not easy to measure the peak ratio in such X-ray energy range as well as to separate gamma/X-ray intensities. The measured ratio of 185.7-keV gamma- ray(^{235}U) and 98-keV X-ray(U) intensity, and ratio of 63.2-keV(^{238}U) and 98-keV(^{235}U) intensities are shown in Fig. 4. As shown in Fig. 4 and 5, the ratio comparing with 98-keV are fluctuate largely. That results from the fitting uncertainty of peak area of 98-keV. This result indicates that the 98-keV peak group is not adequate for unranium enrichment measurement.

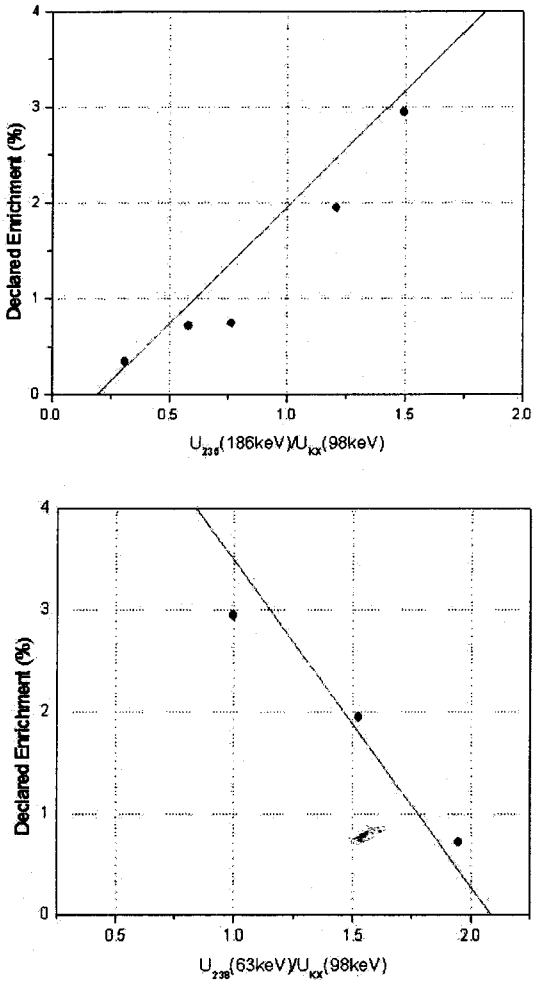

Fig. 4. Intensity ratio curve of the 185.7-keV gamma-ray (^{235}U) and 98.3-keV-ray(U)(Left), and that of 63.2-keV(^{238}U) and 98.3-keV(^{235}U)(Right)

More sensitive and available physical variable is the intensity ratio of 185.7-keV and 63.2-keV. They are pure peak without other interference peaks. Each peaks are proportional to each isotope mass; the 185.7-keV is mass of ^{235}U and the 63.2-keV is that of ^{238}U. Fig. 5 shows the ratio result and the enrichment could be determined within 15% error range. The

commercial package using CZT system shows over 20% error range for low enrichment measurement [3,5,6].

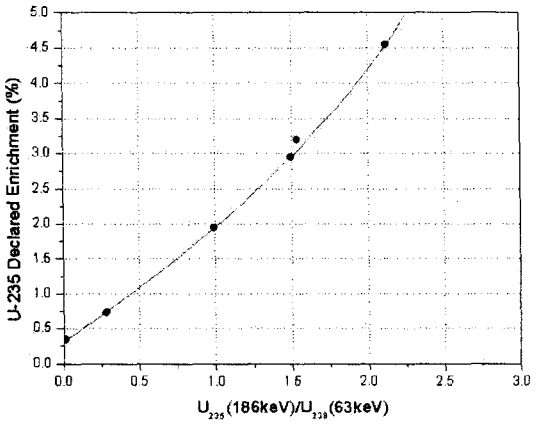

Fig. 5. Measured ratio of 186-keV(^{235}U) and 63-keV(^{238}U) intensities. Solid line is the fitted line by second-order polynomial fitting. Data points deviated from fitting line is the data obtained from unknown samples.

CONCLUSIONS

A uranium isotope enrichment measurement technique by using CZT detector which has good energy resolution near100-keV low energy region was developed. CZT detector characteristics were measured to understand its performance. An energy resolution and an absolute detection efficiency were also determined. The absolute gamma-ray detection efficiency was determined to be less than 1%, and energy resolution was about 4 % near 100-keV energy region.

The uranium enrichment in uranium powders samples was measured by the semi-peak-ratio technique, which used several characteristic gamma-rays related to each isotope. The ^{235}U enrichment in UO2 powder sample could be determine within 15% error range using the newly developed semi-peak-ratio method. This system developed in this study would be used for nondestructive material assay (NDA) dedicated for nuclear safeguards.

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