# $^{230}$TH ASSAY BY EPITHERMAL NEUTRON ACTIVATION ANALYSIS

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$^{230}$Th (Ionium) is an important member of the $^{238}$U decay chain that has recently been shown to be the major contributor to the environmental dose from actinides. (1) Earlier studies (2-4) have shown that following inhalation of $^{238}$U + daughters, the uranium is cleared relatively rapidly from the lung via ciliary action and the gastrointestinal tract, while the $^{230}$Th daughter is removed much more slowly. This produces a "biological enrichment" of $^{230}$Th in later fecal samples and further suggests that $^{230}$Th may be of greater import from the standpoint of dose than heretofore considered.

The lack of a simple, inexpensive, rapid, and sensitive assay method for $^{230}$Th has limited studies of the metabolism and fate of this nuclide within biological systems. The most widely used available methods involve time-consuming, complex, and often tedious chemical separations followed by alpha counting; accuracy and sensitivity of these methods may be wanting, and interferences from uranium, other actinides or isotopes of thorium may further complicate the assay.

The nuclear properties of $^{230}$Th, however, suggest that neutron activation analysis (NAA) may provide a simple, inexpensive, rapid and highly sensitive method of assay. This nuclide has a 1010 barn epicadmium resonance absorption cross-section for activation to 25.52 hour $^{231}$Th; $^{231}$Th has a complex gamma ray spectrum with the most prominent energies being an 84.4 keV complex with a yield of 6.55% (5-6). Although $^{231}$Th emits numerous other photons, yields are significantly (i.e. one or more orders of magnitude) lower, with the exception of a photon at 89.95 keV, which has a yield of 0.95%, or about 1/7 that of the complex line at 84.4 keV.

The suitability of the NAA method in the presence of the $^{238}$U parent and other members of the natural uranium decay chain was examined by activating a quantity of various uranium ores with both a reactor (TRIGA MKI) thermal neutron spectrum having a cadmium ratio of approximately 10. The ore was exposed bare and wrapped in 0.5 mm cadmium sheet to eliminate the subcadmium (i.e. thermal) neutrons. The resultant spectra were counted on a 14% coaxial GeLi detector and showed prominent peaks from

uranium activation and fission products, with a "window" where no (or insignificant) peaks were observed between about 75 and 96 keV. This is in part as expected, for natural activity from $^{231}$Th (daughter of $^{235}$U) is negligible, and would not interfere, and the experiment showed no other potentially interfering peaks. Thus the NAA method appeared feasible as a means of determining $^{230}$Th in the presence of natural uranium.

Pure $^{230}$Th was obtained and irradiated in the reactor shielded by 0.5 mm of cadmium. The resultant spectrum (Figure 1) showed a pronounced broad peak at 84.6 keV, and secondary peaks at approximately 73.5 and 90 keV. Both the 84 and 90 keV peaks are attributable to $^{231}$Th; the other peaks were not identified, but may be attributable to impurities or K x-rays. The presence of $^{231}$Th was verified by observing the decay of the peaks; with the exception of the peaks at 68, 73.5 and 145 keV, all peaks decayed with a half-life of approximately 25.5 hours. A combined sample of $^{230}$Th and uranium ore was also counted together after irradiation. The results clearly showed $^{231}$Th (Figure 2).

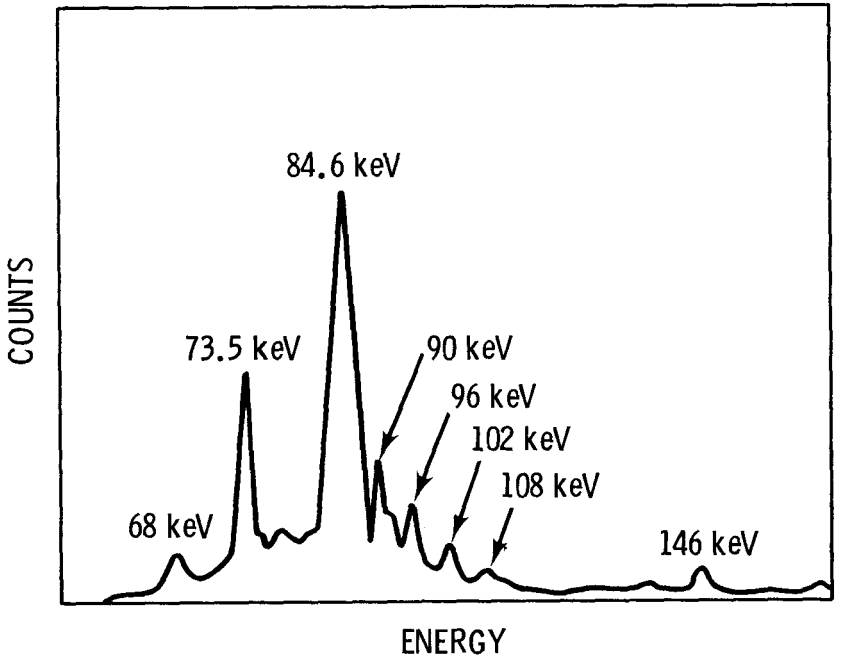


Figure 1. Observed spectrum energy over the range 60 to 150 keV from $^{231}$Th obtained by epicadmium neutron activation of $^{230}$Th.

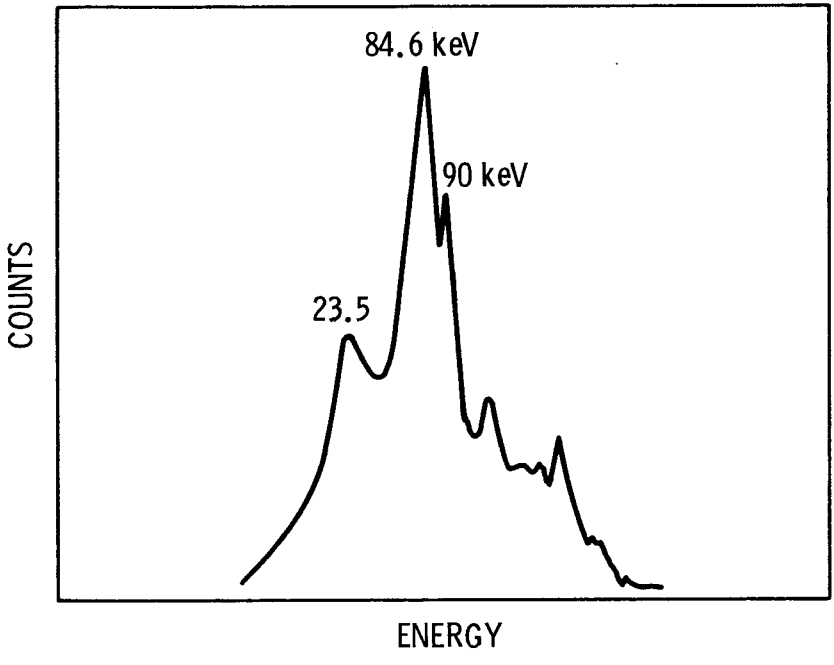


Figure 2. Spectrum in the region of 60-120 keV from mixture of natural uranium and $^{230}$Th following epicadmium neutron irradiation. The peaks identified at 84.6 and 90 keV are from $^{231}$Th; the 73.5 keV peak is from $^{239}$U.

The above preliminary work demonstrates the feasibility of epicadmium NAA as a means of assay of $^{230}$Th, and the data suggest a sensitivity of at least 10 ng for a 10 g sample, 100 minute counting time, and epicadmium fluence of 3 x $10^{12}$n/cm$^2$ (5 x $10^{11}$n/cm$^2$-sec for one minute). While interferences from other substances will be minimized through the use of epicadmium neutrons and a brief post-irradiation delay to permit shortlived activation products to decay, sodium, present in large quantities in feces and other biological materials, may produce significant counts in the low energy channels from Compton scattering of the high energy photons associated with the decay of its activation product $^{24}$Na. The 15 hour half-life of this nuclide is sufficiently close to that of $^{231}$Th to preclude holding the sample for decay. Hence, pre-irradiation sodium removal may be desirable. Further studies along these lines, as well as to improve the method by use of thin intrinsic germanium detectors are now in progress.

REFERENCES

1. Harley, N.H. and Pasternack, B.S. (1979): Health Phys., 37, 291.
2. Stuart, B.O., and Jackson, P.O. (1974): In BNWL-1850, Pt. 1, p. 97.
3. Stuart, B.O. and Beasley, T.M. (1965): In BNWL-122, p. 21.
4. Stuart, B.O. and Jackson, P.O. (1975): In Proc. Conf. on Occup. Health Experience with Uranium, ERDA 93, p. 130.
5. Mughabghab, S.F. and Garber, D.I., (1973): BNL-329, Vol. 1, p. 90-3.
6. Lederer, C.M. and Shirley, V.S., Eds., (1978): Table of Isotopes, Seventh Ed., John Wiley and Sons, N.Y., p. 1418.