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Spectroscopic Compton imaging of prompt gamma emission at the MeV energy range

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Abstract

This work explores a novel tomographic approach to PGAA that is both quantitative and spatially resolved, adapted from a clinical “proton beam range finder” in which MeV gamma rays are imaged by coincidence measurements of Compton scattered gamma rays with multi-detector arrays. We performed preliminary measurements using a Compton camera made with CdZnTe detector arrays on a series of test samples with high-energy (> 1 MeV) gamma emission lines. 3D image reconstructions were performed on the 2.2 MeV peak from H. The image reconstruction methods were also evaluated using the emission data generated by Monte Carlo simulations under ideal conditions.

Keywords

Compton imaging; Prompt gamma activation analysis; 3D elemental distribution

Introduction

Prompt gamma activation analysis (PGAA) is an elemental analysis technique using characteristic gamma rays emitted upon neutron capture by nuclei in a sample [1]. It provides quantitative information for the elemental composition while maintaining the sample integrity. The primary limitation in most applications of PGAA, however, is that it is a bulk technique giving average concentrations, without spatial information. For instance, with standard PGAA, visualization of prompt gamma (PG) signals throughout the sample is achieved by slowly stepping the sample across a small diameter neutron beam to create an elemental map at each position; the gamma spectrum is acquired only at one spot at a time, making this approach a time-consuming process [2].

Compton imaging of gamma-ray sources using a fixed Compton camera (CC) has seen its applications in space telescopes [3] and localization of radioactive sources for homeland

security [4]. The potential of 3-dimensional (3D) PG imaging was recently demonstrated for applications in proton radiotherapy [5, 6]. Thanks to the substantial technological advances in room temperature operated pixelated gamma-ray detectors and fast and miniaturized readout electronics, in addition to more sophisticated reconstruction algorithms, these recent studies have shown the feasibility of imaging PG emissions in the MeV range from elements such C, N, and O [5]. This work explores 3D volumetric Compton imaging of the PG emissions from neutron capture, with the goal of developing a new multidimensional method that can provide both quantitative and spatial information for the elemental distributions within a sample. We report preliminary results from an experiment performed at the PGAA facility at the NIST Center for Neutron Research (NCNR) and computer simulations of Compton imaging during PGAA measurements. From these studies we report on the sensitivity and response of a prototype CC as a function of sample mass and gamma energy and its potential for 3D spectral imaging of individual gamma emission lines.

Theory

The principle of Compton imaging of PG emissions and the reconstruction to localize the source origin is illustrated in Fig. 1. At the MeV energy range, photons have a high probability of undergoing Compton scattering, changing direction and losing energy in the process. By recording the position and the deposited energy of multiple interactions in a pixelated detector, Compton cones can be constructed to reproduce the origin of the gamma ray emission [7]. By using an electronic timing window to identify the coincident events, this method has the advantage of not requiring physical collimation that is difficult to achieve at high energies and not requiring rotating the sample or the detector as in conventional tomography.

For this study, the CORE image reconstruction algorithm [8] is used to generate 3D images based on selected measured gamma emission lines, enabling the identification of an element of interest and its spatial distribution. The CORE algorithm employs an iterative statistical method based on a modified Stochastic Origin Ensemble (SOE) technique [9]. In Compton imaging, individual PGs that scatter in the CC are typically represented by a conic-surface, derived using the physics of Compton Scattering [10], representing the possible locations in space that the PG could have originated from, known as its cone-of-origin. With the CORE (and SOE) algorithm however, each PG is instead represented by a single point on its cone-of-origin. For the reconstruction, a representative point is chosen at random on the section of the cone-of-origin that lies within a pre-defined image reconstruction volume for the first iteration. The reconstruction volume is voxelized and a 3D histogram of the representative points is created as an estimate of the PG emission probability density. For each subsequent iteration a new representative point is chosen on the PG cone-of-origin, and a new emission probability density estimate is calculated each time a new point is chosen for a PG. If the ratio of the new emission probability density estimate (with new representative point) to the previous emission probability density estimate (with previous representative point) is greater than a uniformly distributed random number (U) then the new representative point for the PG is kept and the gamma emission probability density estimate is updated to incorporate the change. If the ratio is not greater than U , the point is moved back to the previous representative point, and the previous emission probability density estimate is used. The

process of selecting random points, comparing them to representative points, and updating the emission probability density estimate is repeated for a preset number of iterations or until an approximately steady state is reached. For full details the reader is referred to [8].

Experimental

The cold-neutron (CN) PGAA instrument at Neutron Guide D (NGD) at the NCNR has a nominal thermal-equivalent neutron flux to $6.5 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$ [11] and radiation shielding designed around a high purity germanium (HPGe) gamma ray spectrometer with the aid of MCNP6 simulations [12, 13]. It is routinely used in non-destructive multi-elemental compositional analysis of bulk material for various applications. The measurement capability is fully reported in a recent comprehensive study [14] on quantifying 100 ppm levels of H in Ti.

The HPGe gamma-ray spectrometer is an n-type, closed-end coaxial detector with 41% relative efficiency (3.05×10^{-5} absolute efficiency at 2.2 MeV) that is equipped with a transistor reset preamplifier. The HPGe detector is inserted vertically into a bismuth germanate (BGO) detector and, in contrast to a CC, it is operated in anti-coincidence mode for Compton background suppression. Data acquisition is performed via a Canberra Lynx digital signal analyzer. The detector assembly views the sample from its side, 90° from the direction of the incident neutron beam at about 60 cm from the neutron beam-sample point of encounter (Fig. 2). The nominal neutron beam is defined by a 2.2 cm diameter ^6Li glass aperture upstream. The sample chamber is lined with ^6Li -loaded silicate glass.

For the present experiment, the CC was placed on the opposite side of the sample chamber at the same detector-to-sample distance of 60 cm, such that simultaneous measurements could be performed. Extra Pb shielding on this side was added to improve the signal-to-noise ratio. The CC was developed based on pixelated CdZnTe detector technology [15] with application specific integrated circuit (ASIC) readout. It has two planes, with 8 modules on each plane. Each module consists of four $2 \text{ cm} \times 2 \text{ cm} \times 1$ (or 1.5) cm CdZnTe crystals and each crystal has a 2D array pixelated anode with 11×11 channels. Each module can provide 3D positional information of the PG interaction as well as energy deposited. The gamma energy spectrum from each module can be summed to form an overall spectrum. 3D images of the PG data measured with the CC were reconstructed using the CORE software.

Results and discussion

Gamma energy spectra were acquired with a series of test samples: 8 mg of NaCl grains, 14 mg Ti foil, and 3 mg parafilm (wax, $\text{C}_n\text{H}_{2n+2}$) disks of 4 mm in diameter. Each sample was irradiated for about 1000 s to 2500 s in the neutron beam while the prompt gamma spectrum was acquired with both the HPGe detector and the CC simultaneously. The sample spectra shown in Fig. 3 give an indication of the energy resolution and efficiency of the camera in comparison with the NIST standard HPGe gamma spectrometer, keeping in mind that the spatial information is obtained from specific energy peaks in the CdZnTe spectrum. The emission lines of Cl, Ti and H shown in HPGe spectrum are also present in the CC spectrum, but the CC spectrum appear to have poorer energy resolution. It is worth noting that the

HPGe detector is heavily shielded by engineering design, whereas the CC is entirely open to the background environment, and therefore the spectrum baseline is expected to be much higher.

Quantitative determination of H mass (proportional to the counts of the H peak) was assessed by incrementing the number of wax disks and integrating the 2.2 MeV peak intensity. First, the spectrum from photoabsorption in the CC is analyzed as in a conventional spectrometer. Figure 4a shows the 2.2 MeV H peak from the CC increases for different number of disks. Figure 4b shows the count rate from each detector as the number of disks increases from 0 to 6. The uncertainties in CC count rates for each disk measurement was less than 1% except for the background (0 disks) which was 3.7%. Both Fig. 4a, b show the observed count rate. The dead time during the measurement on CC was around 3.5%, which means the true count rate is about 4% higher than the observed. From this result, we estimate the absolute efficiency of the CC at 2.2 MeV using the slope of the line in Fig. 4b scaled by the ratio of the solid angle subtended by each detector (estimated to be 3.6 by the ratio of the surface areas, CC to HPGe), giving a value of 1.15×10^{-5} .

Next, tomographic reconstructions of this set of wax disks were performed with the CC data using the PG interactions in the 2.2 MeV emission line of H. Coincident events from a gamma ray that undergoes a Compton scattering, followed by a photoelectric interaction that deposits all its energy, are identified and used for reconstruction as they contain both spatial and initial energy information. The central slices of the reconstructed volumes containing the sample are shown in Fig. 5 for disks 1–6, where the image intensity increase is visually discernible as the number of disks increases. Since the CORE algorithm is a statistical reconstruction method in nature, the reconstruction was run 50 times for each disk number, and the mean of the maximum voxel intensity is compared to the amount of H in different number disks in Fig. 6. It clearly shows a linear dependence, making a calibration of Compton imaging against H content in a sample feasible. The uncertainties in both the mean intensity and the estimated H mass are less than 1%. The linearity also indicates the fraction of 2.2 MeV PGs that undergo a Compton scattering and photoelectric interaction remains constant as H contents increases.

Simulations were conducted using the Monte Carlo code GEANT4 [16] on a series of simple systems to evaluate the feasibility of applying Compton imaging to the element of interest at a spatial scale of interest for elemental analysis. A stack of alternating disks of Ti and H (in the form of water) of 2 cm in diameter and 0.5 mm thick was placed perpendicularly to a set of two detector planes made with CdZnTe material were simulated. PGs produced by a beam of incident neutrons were tracked, and all interactions inside these two planes are recorded. Energy windows centered on the main PG peak of Ti at 1.38 MeV and H emission peak at 2.22 MeV are used to identify coincident events as input to the reconstruction algorithm to recover spatial distribution of each element in the disks. The Ti and H emission images were reconstructed and can be shown separately or as a composite. Figure 7 shows the reconstructed PG emission image as a composite of H and Ti and their profiles interlace in the same fashion as the stacked disks in the original model. The 5.5 mm spacing of the disks is fully resolvable, as are the individual H and Ti disks when each specific emission window is considered separately for the reconstruction.

Conclusions

We have experimentally demonstrated the feasibility of Compton imaging of PG emissions from neutron capture using a set of test samples. Spectra from several samples were acquired using the CC, compared with those from the standard HPGe detector and show good agreement. The H peak was used for tomographic reconstruction. The results suggest that 3D quantitative elemental imaging on the scale of several mm is achievable in simple systems. Monte Carlo simulations were performed for a simple 4-plate stack of H and Ti, the data generated was reconstructed as a case study for an ideal-condition measurement, yielding promising results. Further refinement of the model to reflect experimental setup will be carried out. The result of these studies will facilitate improvement of the reconstruction process, and for future optimized design of the CC for the PGAA application.

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References

1. Lindstrom RM, Révay Z (2017) Prompt gamma neutron activation analysis (PGAA): recent developments and applications. *J Radioanal Nucl Chem* 314:843–858. 10.1007/s10967-017-5483-8
2. Schulze R, Szentmiklósi L, Kudejova P et al. (2013) The ANCIENT CHARM project at FRM II: three-dimensional elemental mapping by prompt gamma activation imaging and neutron tomography. *J Anal At Spectrom* 28:1508–1512. 10.1039/C3JA50162K
3. The Imaging Compton Telescope (COMPTEL). <https://heasarc.gsfc.nasa.gov/docs/cgro/compTEL/>. Accessed 28 Mar 2018
4. MacLeod A (2016) Modular survey spectrometer and compton imager CIRMS conference. NIST, Gaithersburg
5. Polf JC, Rajesh P, Mackin DS et al. (2013) Measurement of characteristic prompt gamma rays emitted from oxygen and carbon in tissue-equivalent samples during proton beam irradiation. *Phys Med Biol* 58:5821–5831. 10.1088/0031-9155/58/17/5821 [PubMed: 23920051]
6. Polf JC, Avery S, Mackin DS, Beddar S (2015) Imaging of prompt gamma rays emitted during delivery of clinical proton beams with a Compton camera: feasibility studies for range verification. *Phys Med Biol* 60:7085–7099. 10.1088/0031-9155/60/18/7085 [PubMed: 26317610]
7. Todd RW, Nightingale JM, Everett DB (1974) A proposed γ camera. *Nature* 251:132–134. 10.1038/251132a0
8. Mackin D, Polf J, Peterson S, Beddar S (2012) Evaluation of a stochastic reconstruction algorithm for use in Compton camera imaging and beam range verification from secondary gamma emission during proton therapy. *Phys Med Biol* 57:3537–3553. 10.1118/1.4767756 [PubMed: 22588144]
9. Andreyev A, Sitek A, Celler A (2010) Fast image reconstruction for Compton camera using stochastic origin ensemble approach. *Med Phys* 38:429–438. 10.1118/L3528170
10. Cree MJ, Bones PJ (1994) Towards direct reconstruction from a gamma camera based on Compton scattering. *IEEE Trans Med Imaging* 13:398–407. 10.1109/42.293932 [PubMed: 18218515]
11. Paul RL, ahin D, Cook JC et al. (2015) NGD cold-neutron prompt gamma-ray activation analysis spectrometer at NIST. *J Radioanal Nucl Chem* 304:189–193. 10.1007/S10967-014-3635-7
12. Goorley T, James M, Booth T et al. (2012) Initial MCNP6 release overview. *Nucl Technol* 180:298–315. 10.13182/NT11-135

13. Turkoglu D, Downing RG, Chen W et al. (2017) A ^3He beam stop for minimizing gamma-ray and fast-neutron background. *J Radioanal Nucl Chem* 311:1243–1249. 10.1007/S10967-016-4954-7
14. Turkoglu D, Chen-Mayer H, Paul R, Zeisler R (2017) Assessment of PGA A capability for low-level measurements of H in Ti alloys. *Analyst* 142:3822–3829.10.1039/C7AN01308F [PubMed: 28952615]
15. Zhang F, He Z, Seifert CE (2007) A prototype three-dimensional position sensitive CdZnTe detector array. *IEEE Trans Nucl Sci* 54:843–848. 10.1109/TNS.2007.902354
16. Agostinelli S, Allison J, Amako K et al. (2003) Geant4—a simulation toolkit. *Nucl Instrum Methods Phys Res Sect Accel Spectrom Detect Assoc Equip* 506:250–303. 10.1016/S0168-9002(03)01368-8

Fig. 1. Principle of Compton imaging with a two-stage pixelated detector. The Compton cones are generated based on the energy and location of the scattered gamma rays to determine the origin of the emission

Fig. 2.

Top view of the NGD-PGAA instrument at the NCNR, with simulated neutron (red) and partial gamma field (green–blue) shown. The Compton camera and the HPGe gamma ray spectrometer are placed on the opposite sides of the sample chamber at about equal distances (60 cm), viewing the neutron irradiated sample.

Fig. 3. Comparison of gamma ray energy spectrum obtained from the CC (black) to that from the HPGe detector (red), for, from top to bottom, NaCl, Ti, wax, and background (with neutron beam on but no sample).

Fig. 4.

a H peak from CC increases with number of disks, **b** H peak count rate from simultaneously measured spectra: count rate in CC versus count rate in the HPGe, as the number of wax disks increases. The data points represent from 0 to 6 disks

Fig. 5.
Reconstructed images of the wax disks. The number of disks in each image is as labeled

Fig. 6.

H peak intensity obtained from tomographically reconstructed images versus H content for the incremented number of wax disks

Fig. 7.

Statistically reconstructed H and Ti images with their spatial profile. The image on the top uses a broad energy window of 2.15–2.3 MeV, which contains both 2.22 MeV H and 2.16, 2.202, 2.206 and 2.24 MeV Ti lines. In the middle row, the energy window is narrowed to 2.21–2.23 MeV, the four peaks are reduced to two, corresponding to H only in the bottom, the energy window is set to around 1.38 MeV from Ti, and the image and the profile show they are located at the correct location