Plastic PTFE Beads Assay

Kris Vorren, Sean MacMullin
Department of Physics & Astronomy, University of North Carolina, Chapel Hill, NC 27599, USA
Triangle Universities Nuclear Laboratory, Durham, NC, 27708, USA
(Dated: March 9, 2011)

INTRODUCTION

Neutron activated plastic beads were γ-assayed in the low background counting (LBC) lab at the Kimballton Underground Research Facility (KURF). Presented in this report are activities from detected isotopes, many of which are neutron activation products of transition metals.

SAMPLE PREPARATION

Plastic PTFE beads were activated on Dec 16th, 2010 at the high flux reactor at ORNL. A sample of 3.0g of this plastic was placed in two glass vials and sent to Virginia Tech. The sample arrived at VT on Jan 19th. While at VT, the sample was removed from the glass vials and placed in plastic liquid scintillator tubes. Due to radiation safety processing delays it was not until Jan 28th that the sample was placed inside the detector for counting. Counts were taken between Jan 28th and Feb 9th, 2011.

MEASUREMENT & ANALYSIS

Measurements were made using the Melissa detector. Melissa is a 50% RE (relative efficiency - to NaI) Canberra LB (low-background) detector. Melissa is in a vertical orientation with a dipstick style cryostat. The lead shield for Melissa consists of 15 cm of Doe Run lead. Inside the lead shield is a 2.54 cm OFHC copper shield to reduce Pb X-rays and Bremsstrahlung from β-decays in the lead shield. The inner cavity is 38cm×38cm×38cm. The FWHM at 1.33MeV is 1.70 keV and the threshold is ∼20 keV [4].

For analysis, a sample spectrum is compared to the background. Background and sample spectra are normalized by dividing out the counting time. Using a library of common gamma lines we then search for peaks that can be used to measure the activity of the sample. For the best results, well-separated peaks with the highest intensities are used. A Gaussian plus a linear background function is fit to each peak used for analysis. The peak area is then extracted by summing the counts in a $\pm 5\sigma$ region for both the sample and background spectra. The fit is used only to obtain the centroid of the peak on the energy axis and to define the limits of the ROI. If a good fit cannot be obtained, for low statistics situations, the ROI is determined using the detector energy resolution. Subtraction of the continuum is then done by defining two regions, one 10σ to the left and the other 10σ to the right of the peak, averaging the sum of the counts in the two regions, and then subtracting from the total integrated peak area. If another peak is between 5σ and 15σ to the left or right, only the region excluding the extra peak is used for averaging. An adjustment factor is multiplied to the integrated continuum area to give an effective range of 10σ for either side. If another peak is within 5σ, then only one side of the peak is integrated for continuum subtraction.

The peak area of a sample or background peak is found by subtracting the continuum from the peak. The net peak area is found by subtracting the peak area of a background peak from the corresponding sample peak. If there is no peak on the background spectrum, the background peak area is set to zero.

The activity of a specific peak is then given by:

$$A_\gamma = \frac{\text{Net Peak Area [cps]}}{\epsilon_\gamma \, m} \tag{1}$$

where ϵ_γ is the peak efficiency, and m is the mass of the sample.

If no statistically significant peak is present, an upper limit can be placed on the activity in a sample. The signal in an energy bin is then required to be less than or equal to the upper limit, 90% C.L. - 1.64σ (σ here is just the square root of the recorded counts) [1].

Peak efficiencies are determined by using a detailed Monte Carlo simulation in `MaGe` [2]. Once a detailed sample geometry has been coded into the simulation, it is doped uniformly with isotopes of interest, usually from the ^{238}U, ^{232}Th, and ^{40}K decay chains, along with any others that may be present in the sample, e.g. ^{60}Co. The primary γ-rays from these decays are tracked from the emission of the source to absorption in the detector active region. The main advantages to using a pure Monte Carlo simulation is that self-attenuation in the sample is accounted for, and there are no limitations on source or detector configurations.

The energy deposits in the simulated spectrum are convolved with the finite energy resolution of the detector. The resolution factor, σ_{res}, has been measured as a function of γ-ray energy (E_{keV}) from 303–1836 keV using radioactive point sources. Using the following equation from [3], these data were fit to

$$\sigma_{res} = \sqrt{a^2 + b^2 E_{keV} + c^2 E_{keV}^2} \tag{2}$$

The net peak area in the simulated spectrum is determined by fitting the peak of interest with a Gaussian and subtracting a linear background. A correction term must also be included to account for systematic error in the Monte Carlo. For a sample in Melissa located near the top of the detector, f = 1.04±0.06 [4]. The absolute peak efficiency, including branching ratios, can then be determined from

$$\epsilon_\gamma = f \frac{\text{Net Peak Area}}{\text{Number of Events Simulated}} \tag{3}$$

RESULTS

A list of γ-active isotopes found in the spectrum is given. All of them had activities that dominated anything seen in the background. All of the isotopes found were products of neutron reactions. Listed are the time averaged activities and extrapolated activation activities. The activation activity was determined from

$$A_0 = \frac{-\lambda \bar{A} \Delta t}{e^{-\lambda t_i}(e^{-\lambda \Delta t} - 1)} \tag{4}$$

where A_0 is the extrapolated initial activity, $\bar{A}$ is the measured time averaged activity, t_i = 43 d is the time between activation and counting, Δt = 11.75 d is the counting time, and λ is the decay constant, i.e $\ln(2)/t_{1/2}$.

Detected Isotopes

^{233}Pa: $t_{1/2}$ = 27 d, $\bar{A}$: 15.4 ± 1.4 Bq/kg (590 ± 50 ppb ^{237}Np).
Measurement of the ^{233}Pa line at 312 keV. Note that the value given for the ^{237}Np assumes secular equilibrium.

^{22}Na: $t_{1/2}$ = 949.7 d, $\bar{A}$: 14.7 ± 0.7 Bq/kg, A_0: 15.3 ± 0.7 Bq/kg.
Line at 1275 keV.

^{46}Sc: $t_{1/2}$ = 83.79 d, $\bar{A}$: < 40 Bq/kg, A_0: < 57 Bq/kg.
Two lines were found at 889 and 1120 keV. There was a large discrepancy between the activities measured from the two lines. At 889 keV, 12.7 ± 1.2 Bq/kg was measured, while at 1120 keV, 37.2 ± 1.4 Bq/kg was measured. The ^{214}Bi background at 1120 keV was taken into account.

^{51}Cr: $t_{1/2}$ = 27.7 d, $\bar{A}$: 10.64 ± 0.02 kBq/kg, A_0: 36.03 ± 6 kBq/kg.
Line at 320 keV.

^{54}Mn: $t_{1/2}$ = 312.4 d, $\bar{A}$: 7.2 ± 0.9 Bq/kg, A_0: 8.0 ± 1.0 Bq/kg.
Line at 835 keV.

^{59}Fe: $t_{1/2}$ = 44.5 d, The 1099 and 1291 keV line resulted in an activity measurement of $\bar{A}$: 272 ± 3 Bq/kg and A_0: 582 ± 6 Bq/kg while the 192 keV line resulted in $\bar{A}$: 255 ± 14 Bq/kg and A_0: 545 ± 3 Bq/kg.

^{58}Co: $t_{1/2}$ = 70.86 d, $\bar{A}$: 9.7 ± 0.8 Bq/kg, A_0: 15.7 ± 1.3 Bq/kg.
Line at 811 keV.

^{60}Co: $t_{1/2}$ = 1923.55 d, $\bar{A}$: 1.522 ± 0.004 kBq/kg, A_0: 1.549 ± 0.006 kBq/kg.
These measurements were based off the lines at 1173 and 1333 keV respectively. The coincidence peak at 2505 keV and single and double escape peaks at 822 and 311 keV were also found.

^{65}Zn: $t_{1/2}$ = 244.26 d, $\bar{A}$: 102 ± 3 Bq/kg, A_0: 117 ± 3.
Line at 1115 keV.

^{124}Sb: $t_{1/2}$ = 60.2 d, $\bar{A}$: 95 ± 9 Bq/kg, A_0: 167 ± 3 Bq/kg.
Averaged value of the activities measured from the 602, 1368, 1690, and 2091 keV lines. The activities of individual lines were all within 3σ of each other.

^{95}Nb: $t_{1/2}$ = 34.975 d, $\bar{A}$: 2.2 ± 0.8 Bq/kg, A_0: 6 ± 2 Bq/kg.
Line at 766 keV.

^{185}Os: $t_{1/2}$ = 93.6 d, $\bar{A}$: 5.4 ± 0.9 Bq/kg, A_0: 7.8 ± 1.3 Bq/kg.
Line at 646 keV.

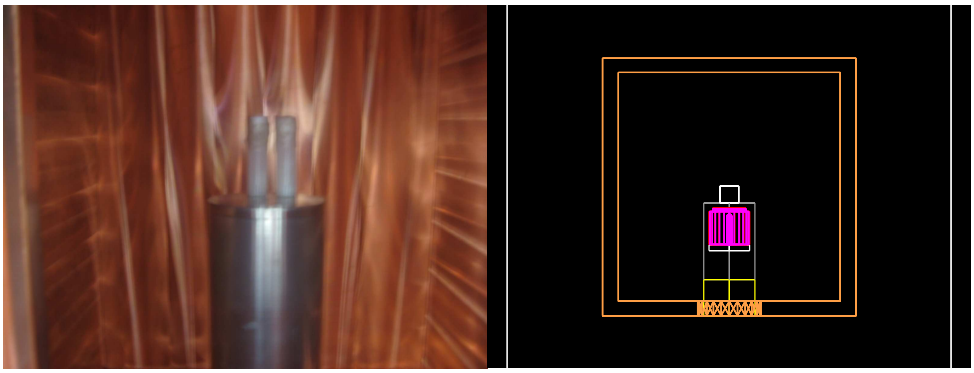
^{192}Ir: $t_{1/2}$ = 73.83 d, $\bar{A}$: 9.2 ± 1.2 Bq/kg, A_0: 14.5 ± 1.9 Bq/kg.
Activity of the 468 keV line. The 316 keV line was also found, however due to the presence of the 320 keV ^{51}Cr and the 312 keV ^{233}Pa line, it was not possible to accurately measure the activity of the line.

CONCLUSIONS AND REMARKS

Many neutron activation products were found in the beads spectrum. Most of the products were isotopes of transition metals, probably from stainless steel. Some of the lines were strong enough that single escape, double escape, and addition peaks were also seen. Examples include 822, 311, and 2505 keV–all from ^{60}Co. Because the sample was not assayed until over a month after preparation, it is likely that there were some products which decayed away before the sample was even placed in the detector.

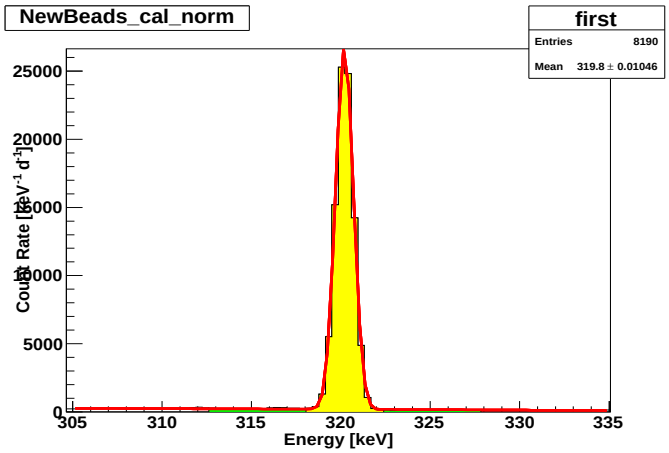
[1] G. Gilmore, *Practical Gamma Ray Spectrometry* (2008), 2nd ed.
[2] M. Bauer et. al, Journal of Physics: Conference Series **39** (2006).
[3] G. F. Knoll, *Radiation Detection and Measurement* (Wiley & Sons, Inc., Hoboken, NJ, 2010), 4th ed.
[4] P. Finnerty et. al, arXiv:1007.0015v1 (2010).

FIGURES



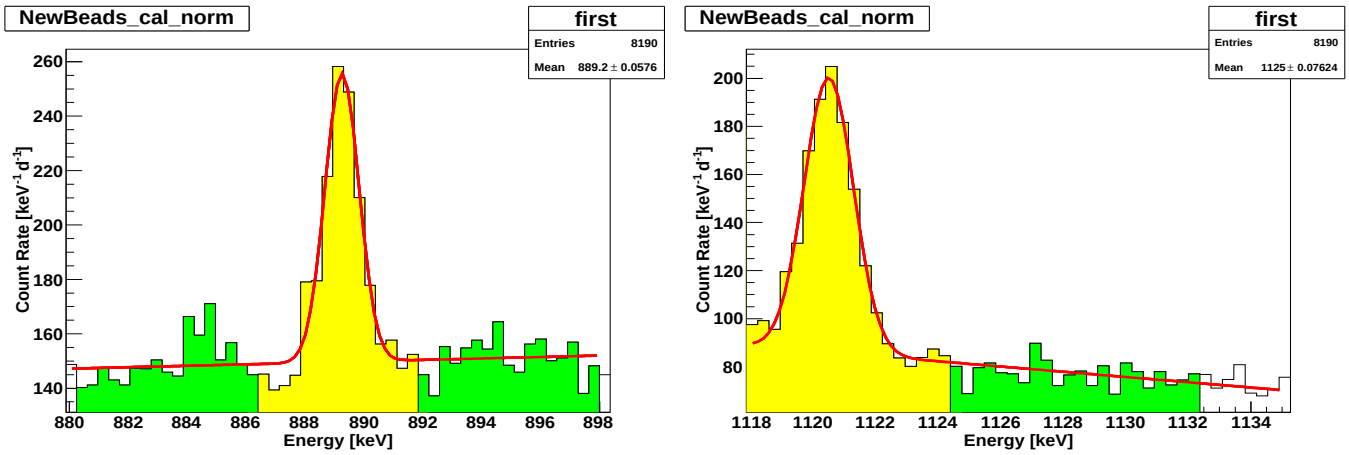
(a) Sample (b) Simulated Geometry

FIG. 1: Geometry of the assayed sample. For simplicity, the sample was approximated as a point source for the MC.



(a) Plastic Beads

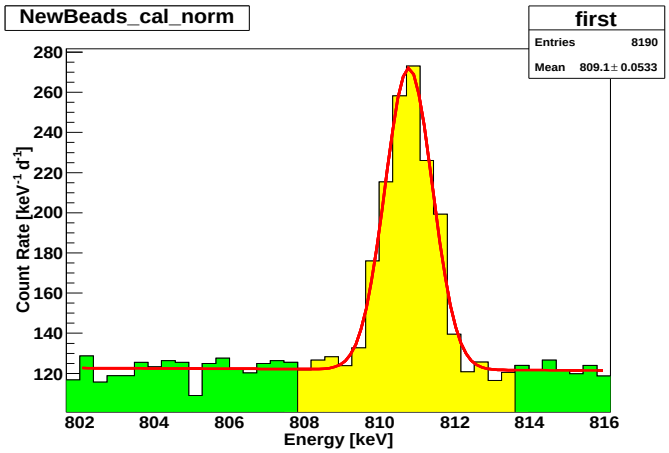
FIG. 2: 320 keV γ line from ^{51}Cr. Yellow is the integrated peak, green is the region of subtracted continuum.



(a) 889 keV

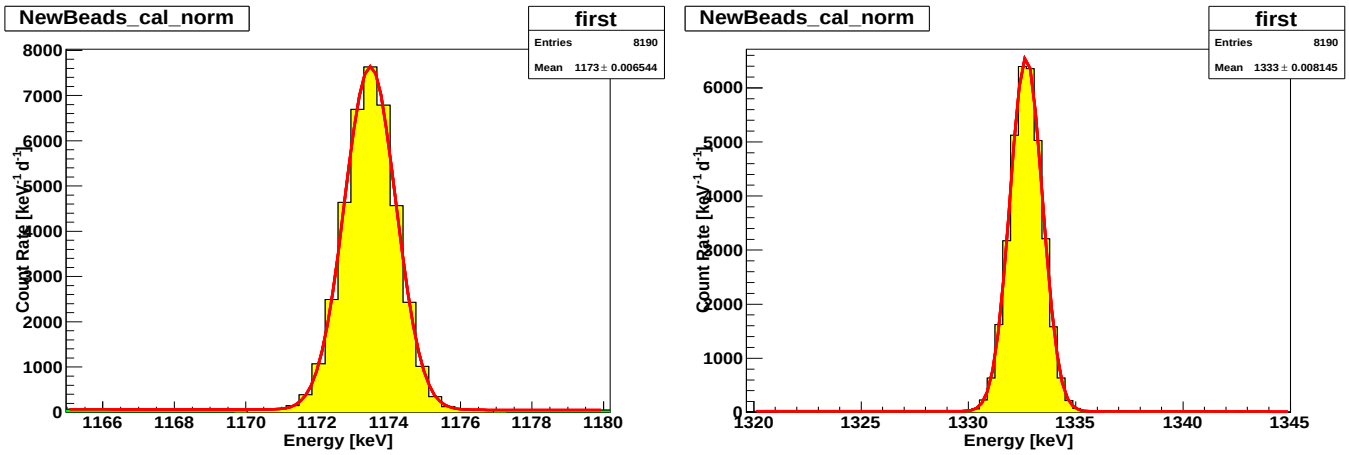
(b) 1120 keV

FIG. 3: ^{46}Sc. There was a line at 1115 keV, so only the right side was used for continuum subtraction in fig 2b.



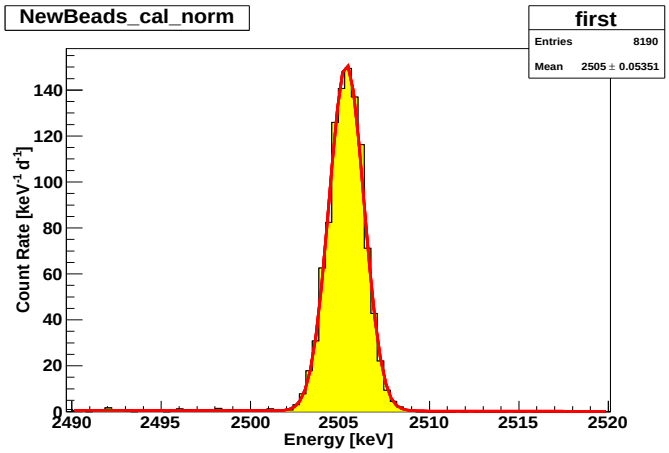
(a) 811 keV

FIG. 4: ^{58}Co.



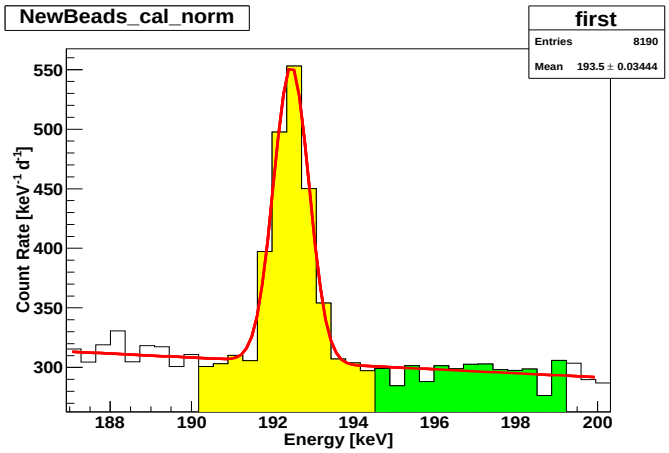
(a) 1173 keV

(b) 1332 keV

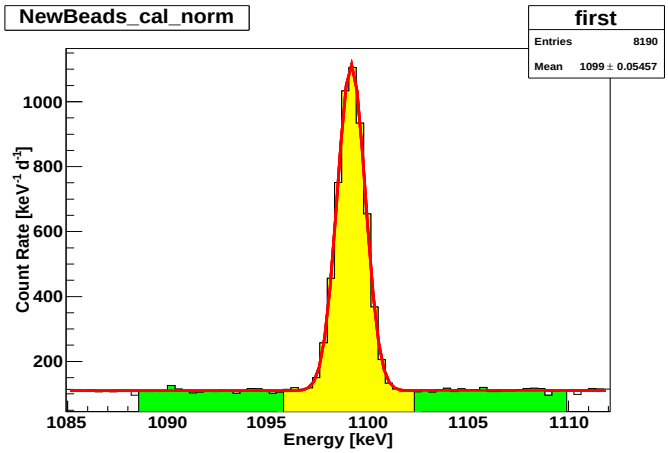


(c) 2505 keV addition line

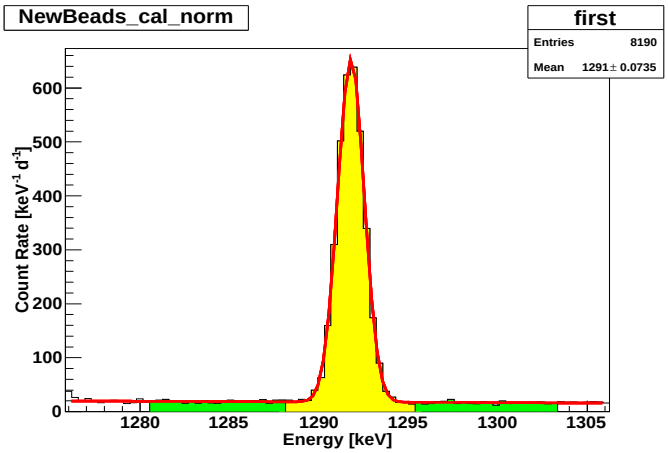
FIG. 5: ^{60}Co.



(a) 192 keV

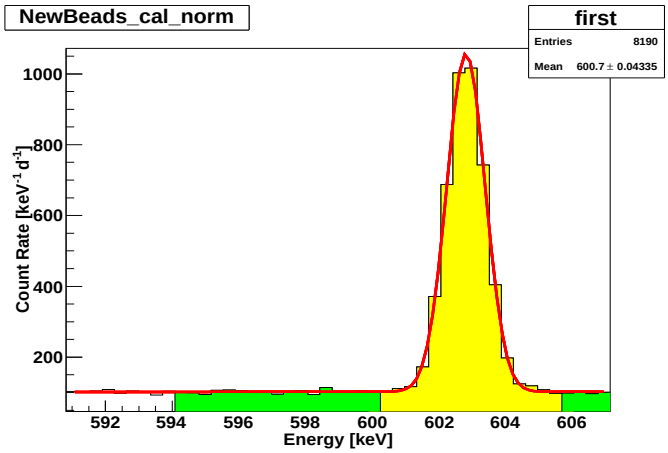


(b) 1099 keV

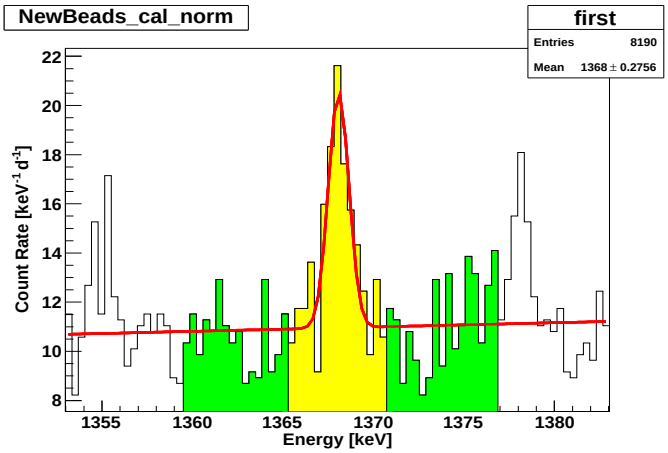


(c) 1291 keV

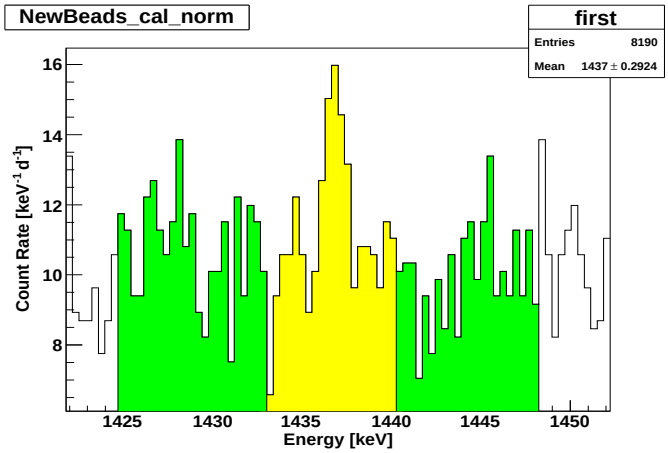
FIG. 6: ^{59}Fe.



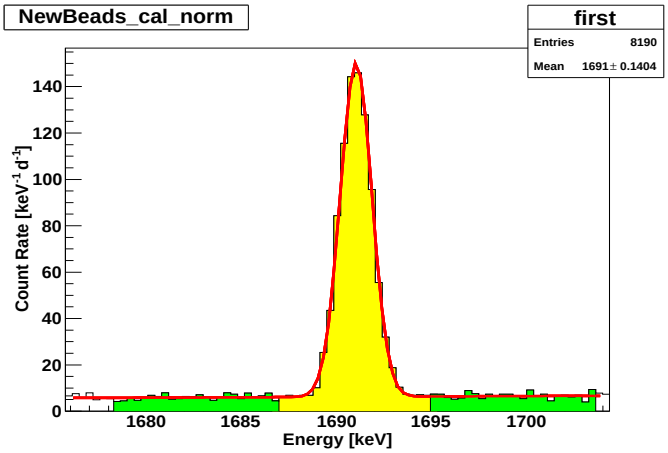
(a) 603 keV



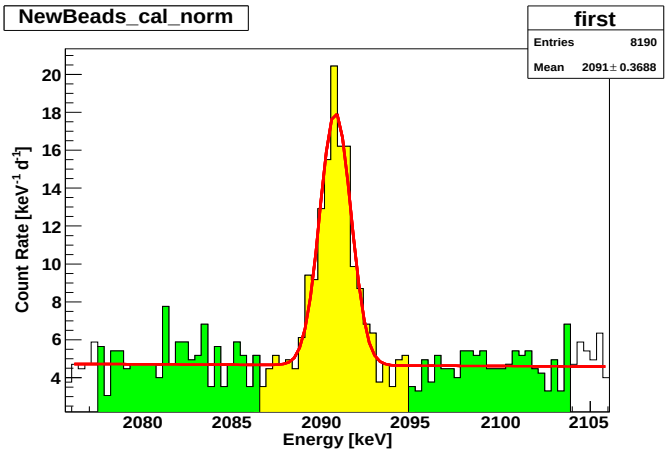
(b) 1368 keV



(c) 1437 keV

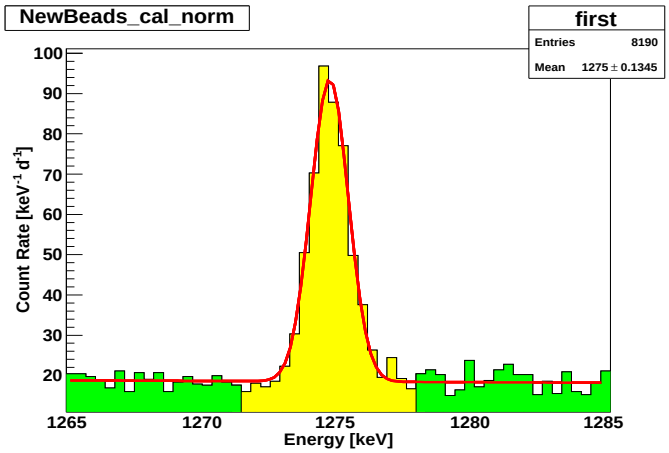


(d) 1691 keV



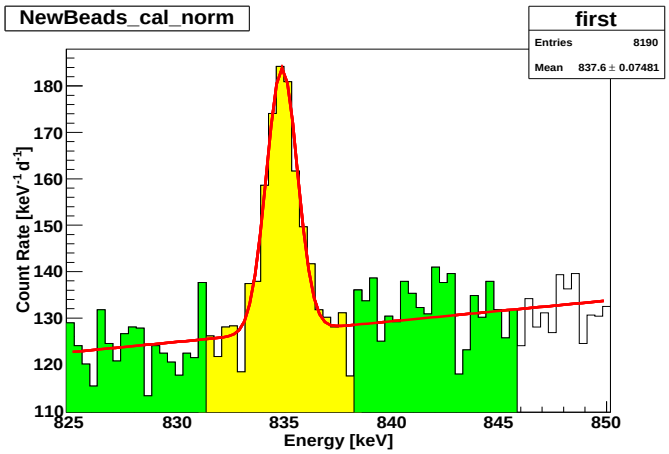
(e) 2091 keV

FIG. 7: ^{124}Sb.



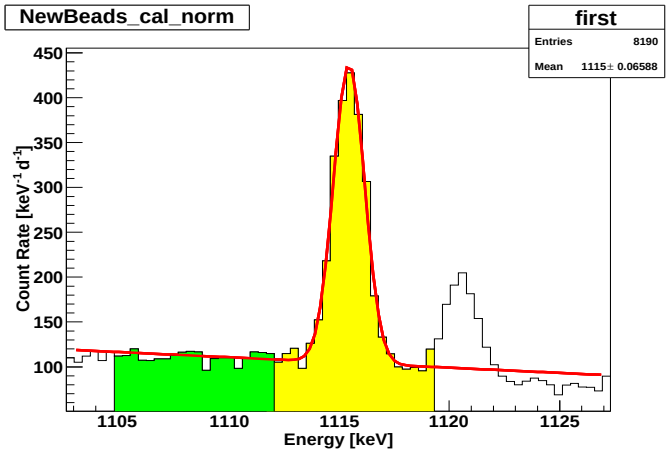
(a) 1275 keV

FIG. 8: ^{22}Na.



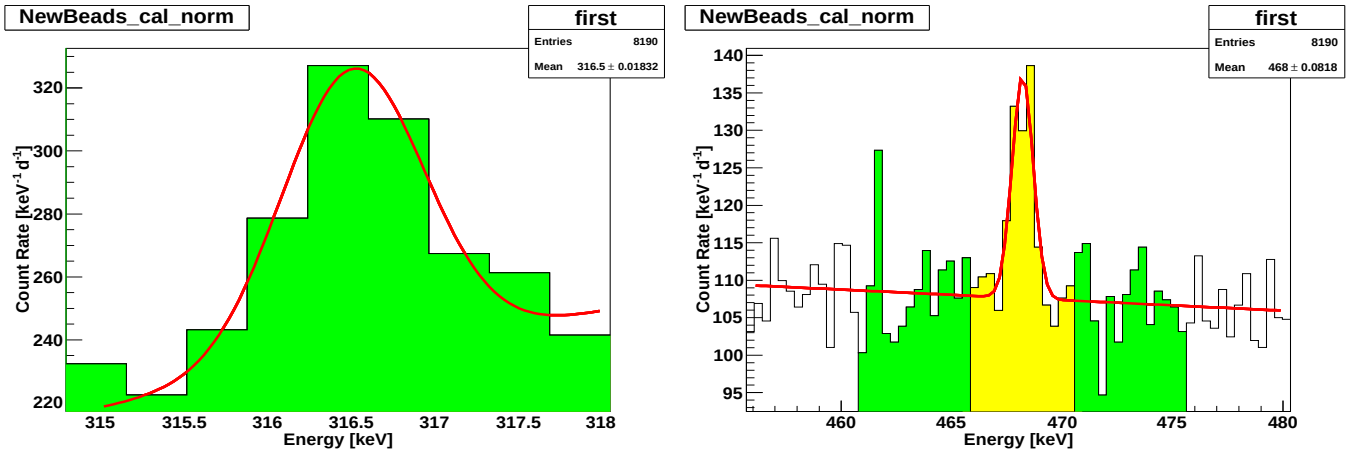
(a) 835 keV

FIG. 9: ^{54}Mn.



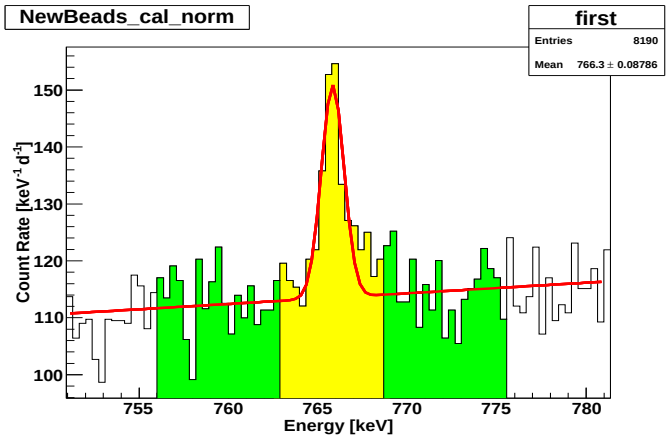
(a) 1115 keV

FIG. 10: ^{65}Zn.



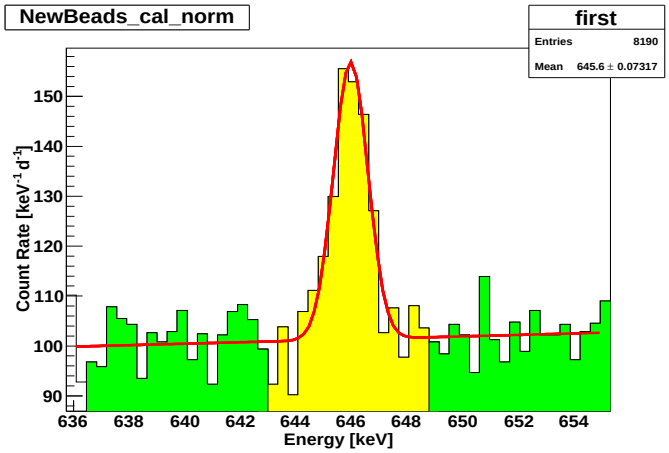
(a) 316 keV (b) 468 keV

FIG. 11: ^{192}Ir. There was a line at 1115 keV, so only the right side was used for continuum subtraction in fig 2b.



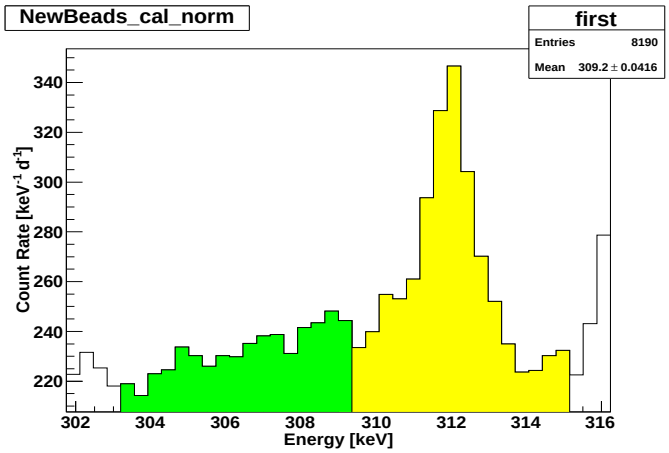
(a) 766 keV

FIG. 12: ^{95}Nb.



(a) 646 keV

FIG. 13: ^{185}Os.



(a) 312 keV

FIG. 14: ^{233}Pa.

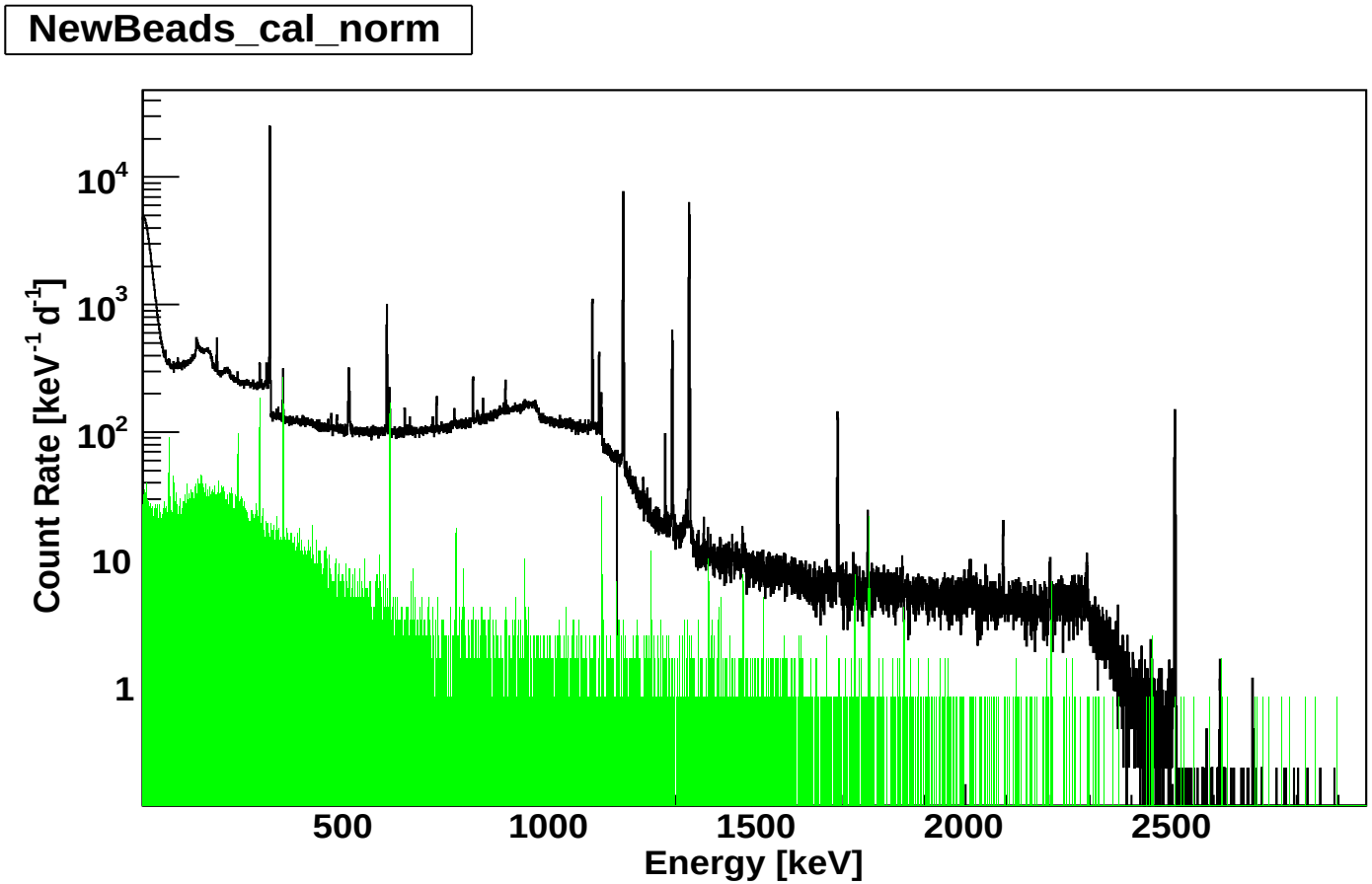


FIG. 15: Sample and background spectra for the second plastic bead sample. Black is the sample, green is the background. The counting live time for the sample and background were 11.75 days and 3.25 days respectively.