Cf-252 Spontaneous Fission

Navrit Bal

College of Letters and Science
University of California at Berkeley
Berkeley, California, U.S.A.

Nuclear Engineering 102
navrit@berkeley.edu

April 5, 2015

Abstract

Our experiment confirms that Californium-252 (Cf-252) undergoes spontaneous fission, releasing predominantly alpha partciles ($6.26 \pm 0.15 MeV$) with a wide range of 'daughter' nuclei (fission fragments) with their associated energies ($\approx 5 - 120 MeV$). We found how the energy of the fission products is reduced by 39-50% by passing through gold foil. The thickness of the goil foil was calculated to be $116 nm$ using the Bethe Formula along with the shift in alpha peak energy (with and without the foil). The expected count rate was estimated to be slightly higher than the measured count rate of the Cf-252 source alone.

Note: British English spelling is adhered to within this document.

1 Theory

Fission is the nuclear process whereby a nucleus of an atom decays to form at least 2 'daughter' products. These are a combination of lighter nuclei and other nuclear particles, primarily neutrons. Most nuclei require additional energy in order to undergo a fission reaction, not all do. Some nuclear isotopes are sufficiently massive so that they undergo spontaneous fission; nuclei can decay via fission with no additional energy input, such as Cf-252.
The decay products of these spontaneous fissions are not evenly split by mass as one may expect. The decay products have a range of masses distributed across at least 2 products with a sum of the parent nucleus. Cf-252 undergoes spontaneous fission for 3.09% of decays; Cm-248 is the alpha decay product of Cf-252 which occurs for 96.91% of decays.

Cf-252 spontaneous decay products mass can theoretically be between 1 and 251 A.M.U. (atomic mass units). There is a double peak distribution from the nucleus splitting in an asymmetric way. The humps are not soothly distributed due to the stability of nuclei with even/odd numbers of neutrons/protons.There is a dependence between fissile nucleus energy and the symmetry of the distribution. The higher the energy of the nucleus, the more symmetric the distribution will be.

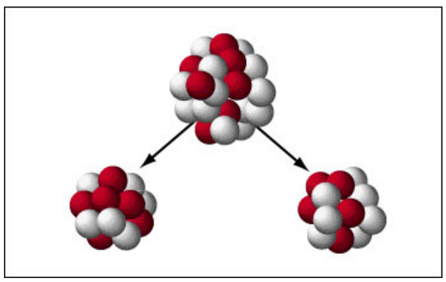

Figure 1: Diagram showing spontaneous fission with 2 daughter nuclei and importantly, no incoming particles (neutrons).

2 Experiment

Spontaneous fission is a special case of a fundamental nuclear decay process. This phenomenon can be explored with only the sample and not a nuclear reactor or accelerator and is therefore inherently more safe. This experiment is able to identify asymmetric mass distribution of fission decay fragment products from Cf-252 - 2 overlapping Gaussian (humps) shape.

The experiment shows how fission fragments (charged particles) are affected as they pass through a thin gold foil with minimal resistance - the apparatus operates in a vacuum chamber (10-30 mTorr). We can quantify what is a normal background reading for that system. We can also show how activated (very dry) charcoal can gain radioactive fission fragments from the Cf-252.

2.1 Method and Procedure

1. Measure gamma emissions from Cf-252 fragments with and without gold foil between the Si (silicon) detector and source. Measure distance between source and detector and compare between measurements.

2. Acquire or find a background gamma spectrum measurement for comparison with measurements with samples.

3. Bonus: measure gamma spectrum of the Cf-252 with the HPGe detector

4. Bonus: measure gamma spectrum of the activated sample which has been stored next to the Cf-252

2.1.1 Electronics, experimental diagrams

1. ORTEC surface barrier silicon (Si) detector [1]

2. Coaxial HPGe semiconductor detector (High Purity Germanium)

3. Californium-252 radioactive source (Cf-252) - 96.91% alpha emitter and 3.09% spontaneous fission. Activity was 4225Bq as measured on 1 November 2011.

4. Vacuum chamber - a lower ring sealed with a rubber ring

5. Vacuum pump and tubing equipment

6. High Voltage (HV) module operated at 50V (Dual solid state detector bias supply)

7. Pre-amplifier

8. ORTEC Linear amplifier

9. ORTEC Easy MCA (Multiple Channel Analyser)

10. MAESTRO - Computer

11. Borated polyethylene - neutron shielding (green)

12. Black cloth

13. Calliper

14. Gold Foil (unknown thickness)

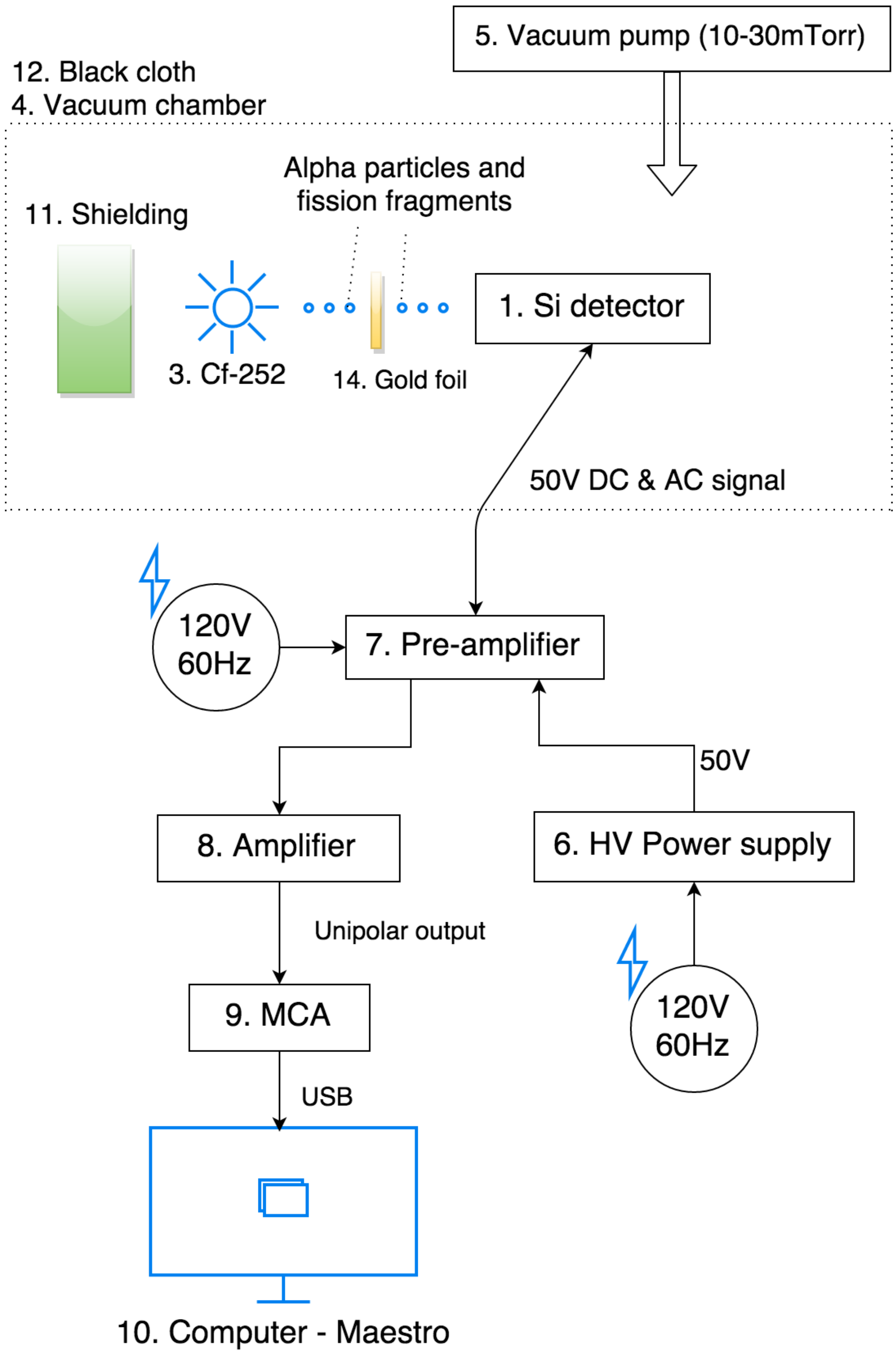

Figure 2: This is a logical overview diagram of the experimental apparatus.

2.2 Data

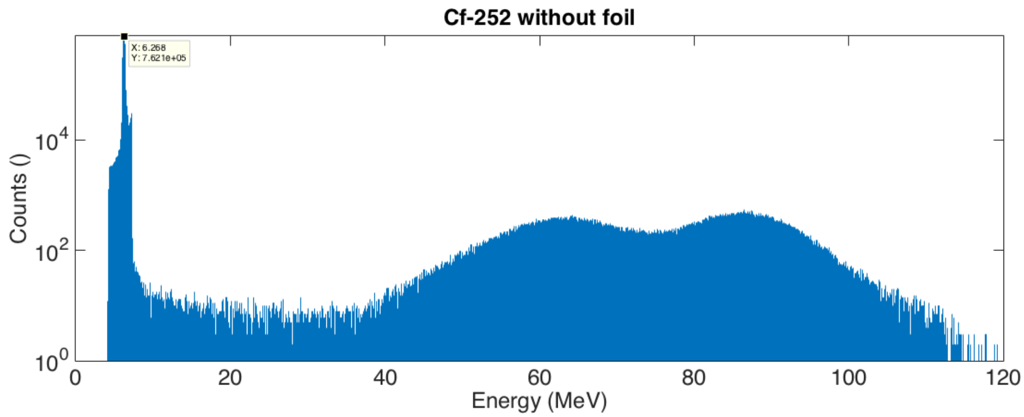

Figure 3: As expected the peak energy of the alpha particles is $6.26 \pm\ 0.15 MeV$, with the remaining 2 Gaussian (assumed) peaks centred around 64 and 88 MeV. These correspond to a range of isotopes mostly between Z=66 to 172 as shown in [4]

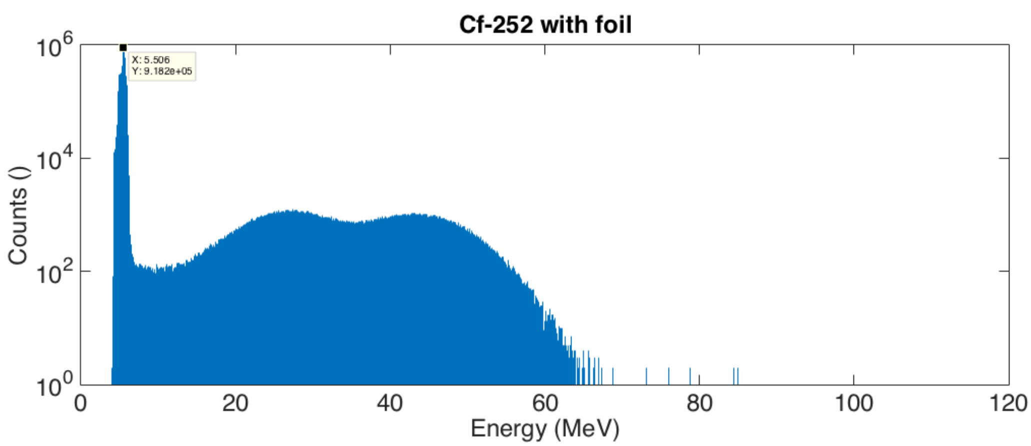

Figure 4: The alpha peak was shifted by approximately 0.7 MeV (around 11%) to $5.51 \pm 0.15 MeV$. The other 2 Gaussian (assumed) peak centres were reduced to 25 and 45 MeV - $39 - 50\%$ of the measurement with foil.

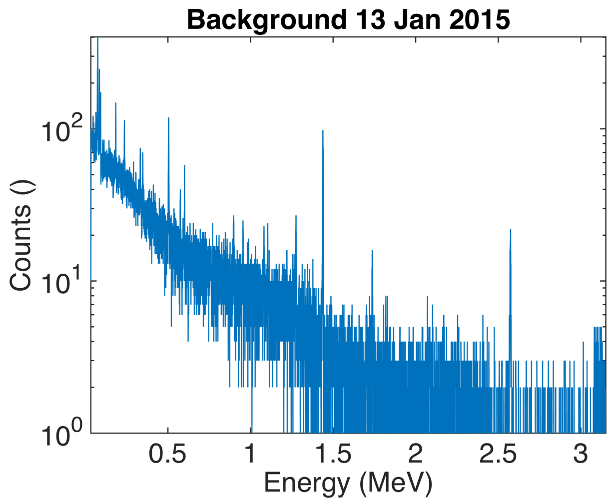

Figure 5: A supplied background measurement - as expected, this shows how there is a very low background contribution as this measurement was taken over 8.3 hours.

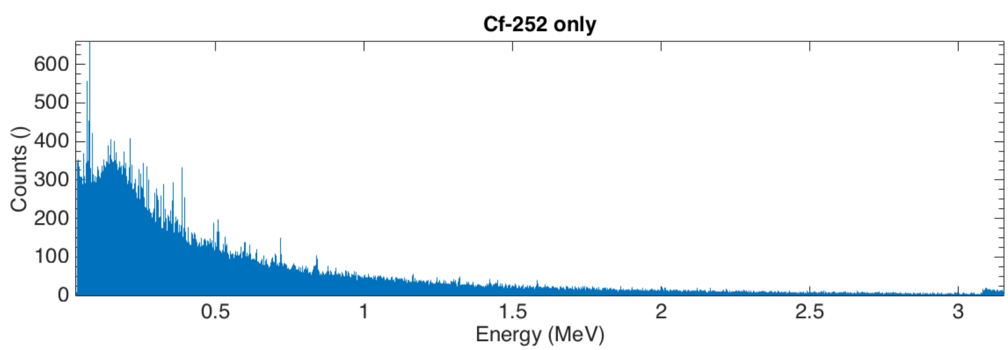

Figure 6: This peaks in this plot show what energies were detected when we placed the Cf-252 source into the detector alone. The largest 2 peaks at 75.8 and 67.7MeV do not appear to correlate to any Cf-252 emission lines. Any peaks above 155KeV are not attributable to Cf-252 according to the latest LBNL ENSDF entry [3]

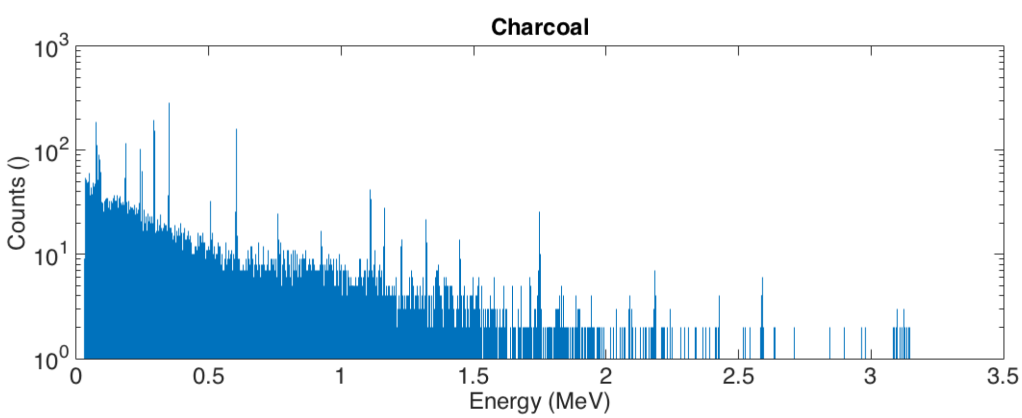

Figure 7: These peaks correspond to a wide range of possible daughter nuclei as shown comprehensively in the LBNL ENSDF text file[2]. The peaks have not been labelled here for plotting and readability reasons.

2.3 Analysis and Results

The Cf-252 source alone in the chamber seems to reveal that there are significant emissions from other sources than the Cf-252. This is assuming that the calibration was correct. The gamma energies which are expected according to [3] are below what our system is able to measure - the gamma anergies expected from Cf-252 are $43.38 \pm 3, 100.4 \pm 3$ *and* 155.0 ± 4 KeV.

Activated charcoal has been shown to have fission fragments embedded within it. This can be viewed in analogy in the same way that ions can be embedded within semiconductors in the doping process. We can confirm the presence of these fission fragments by identifying gamma spectrum peaks which are associated with their respective decay modes. See figure 7 for the spectrum for the activated charcoal and figure 6 for the background measurement.

Figure 7 shows the manually identified peaks in the activated charcoal spectrum (Figure 7). The way that I determined whether a peak was actually a peak was if the maximum point was at least 2 times that of the value 3 bins either side, this was done manually.

Measured Energy (KeV)	Gamma energy (KeV)	Isotope
350.0 0.4	349.6 2	118Pd
293.9 0.4	294.0 3	149Ce
75.8 0.4	75.9 5	76Zn
604.7 0.4	604.7 2	134Cs
241.2 0.4	241.5 2	155Er
186.2 0.4	186.3 1	94Kr
1110 1	1109.4 4	80Ga
762 1	762.3 2	96Pd
1749 1	1748.4 1	149Er
1163 1	1163.6 3	122m In
1321 1	1321.1 1	136I
925.5 0.4	925.6 1	121In
1227 1	1226.0 2	131Sn
1447 1	1446 0	92Br
2183 1	2182.4 2	94Sr
2590 1	2590.0 5	155Nd

Table 1: Charcoal possible daughter decay nuclei with associated uncertainties for the least significant digit after each value. The rows (isotopes) are ordered in terms of descending counts as seen in Figure 7. This table only shows a few of the possible isotopes that these energies correspond to - there are many possible isotpes that fit into these enrgies given their respective uncertainties.

The Bethe Formula (relativistic) will now be used to show the energy loss (MeV) per micrometer (micron). The formula in use is as follows with some of the variables defined below:

$$-\frac{dE}{dx} = \frac{4\pi}{m_e c^2} \left(\frac{e^2}{4\pi\epsilon_0}\right)^2 \frac{nz^2}{\beta^2} \left[ln\left(\frac{2m_e c^2 \beta^2}{I(1-\beta^2)}\right) - \beta^2\right]$$

$$n = \frac{N_A Z \rho}{A M_U};\ n_{Au} = 4.6608 \times 10^{30} m^{-3}$$

$$\beta = \frac{v}{c};\ c\ [ms^{-1}] -\ speed\ of\ light;\ I\ [eV] -\ mean\ excitation\ energy;$$

$$\beta^2 = (\frac{v}{c})^2 = (\frac{\sqrt{c^2 - \frac{mc^4}{E_\alpha + 1}}}{c^2})^2;\ \gamma = 3.5 \times 10^6$$

$$I_Z = 10eV \times Z;\ I_Z = 790eV$$

Substituting the above values into the Bethe formula leads to the following answer:

$$-\frac{dE}{dx} = 53.8 MeV \mu m^{-1}$$

Using this value, I estimate that the gold foil through which the alpha particles were attenuated had approximately the following thickness:

$$Thickness\ of\ Gold\ Foil,\ x = |E_\alpha \div -\frac{\Delta E}{\Delta x}| = |6.26MeV \div -53.8MeV \mu m^{-1}| = 116nm$$

A point to note is the actual activity of the Cf-252 sample was not the same at the time of using it as it was on 1 November 2011. The sample was mainly on 12 March 2015 (1228 days later), and so a calculation of the estimated count rate on the day of primary usage is now presented with this information. The half life of Cf-252 is 6.217 years ($t_{\frac{1}{2}} = H = 2271\ days$).

$$A = A_0\ 0.5^{\frac{t}{H}}$$

Where A = current activity, A_0 = activity at time t, t = time since reference activity (1228 days) and H = the half life (2271 days)

$$A = 4225\ Bq\ 0.5^{1228\ d/2271\ d}$$

$$A = 2904\ Bq$$

Assuming a detector efficiency of 1, activity frequency $A = 2904Hz$ and solid angle per detector ($\frac{\pi R^2_{Detector}}{4\pi R^2_{Distance}}$), $R_{Detector} \approx 3in,\ R_{Distance} \approx 10in\ = \frac{9}{400}$ - the approximate expected number of gamma detection events ($\gamma_{expected}$) can be estimated as follows.

$$\gamma_{expected}\ (Frequency\ of\ seeing\ any\ particles\ from\ Cf{-}252) = (\frac{5^2}{4 * (23 \pm 5})^2 {*} 1 {*} 2904Hz \approx 40{\pm}15Hz$$

The average count rate of the Cf-252 with no foil was determined by dividing the total counts by the total (live) time that the detector was running for as follows.

$$Average\ count\ rate = \frac{Total\ counts}{Total\ live\ time} = \frac{3105130}{169214} \approx 18Hz$$

Comparing this estimated frequency to the measurement taken using Cf-252 and no foil, we find that the expected count rate ($40 \pm 15Hz$) is slightly high than the no foil measurement ($\approx 18Hz$) but within the same order of magnitude.

2.4 Error Analysis

To confirm that the preset calibration was accurate, we compared the known alpha particle energy to the measured energy as found by our apparatus. The measured alpha energy was $6.26 \pm 0.15 MeV$ compared to the NNDC, BNL (National Nuclear Data Center, Brookhaven National Laboratory) value of $6.11810 MeV(81.5\%)$ *and* $6.07564 MeV(14.5\%)$. This gives experimental validation considering the NNDC value falls within our measured value with our associated uncertainties.

Each channel (bin) covered 150 KeV and so that was the main limitation in determining the accuracy of the measured energies. Using 2 systems (different MCAs) led to one system with 2048 channels and the other with 8192 channels.

The inherent statistical uncertainty of $\sqrt{N}$ was the major source of uncertainty for the count number. It was not plotted on any graphs due to the types of features that we are highlighting. The features of interest are shapes of distributions and how they change and or at what energy the peaks/humps (overlapping Gaussians) appear.

3 Conclusions

Our measurements show how the energy loss of the primary alpha peak is approximately 0.7MeV due to the addition of the gold foil. We have also demonstrated the energy loss of the Cf-252 fission fragments with figures 3 and 4 - the whole gamma spectrum is compressed down as the higher energy fragments lose higher proportions of their energies compared to lighter particles.

The use of the Bethe formula gives the energy loss ($\frac{-dE}{dx}$) that the alpha particle experiences as it passes through the gold foil (about 0.7MeV). I used that value to estimate the thickness of the gold foil at $1.16nm$.

I estimated the expected count rate of gammas using the geometrical properties of the system to show how the projected solid angle of the detector from the source would reduce the detected count rate. This calculation led to finding that the expected count rate was slightly higher than the calculated count rate, although very much within the same order of magnitude. Presumeabley this means that all of the major factors were geometrical as expected.

The various possible isotopes that the Cf-252 source spontaneously decays into have been referenced in [2]. This shows the full distribution of isotopes that this fission reaction leads to - a large number. This is why I displayed only some of the possible matches with the measured energies from the charcoal, these isotopes are not necessarily what caused those emission lines. The resolution of the system is not sufficient to determine which isotope(s) caused those specific energies.

Ways to improve the experiment include simulations to obtain expected values (I could not run the necessary software), have longer recording times, have more shielding (ideally background radiation would not exist), measuring the activity of the source instaed of calculating it and having a higher resolution so that gamma peaks can be more accurately determined.

4 References

[1] on page 1 - http://bit.ly/1brhUpj - accessed on April 5, 2015

Figure 1 - http://bit.ly/1FEHNie - accessed on April 5, 2015

Table 1 - Nuclear Science Division LBNL - Data compiled from http://ie.lbl.gov/toi/radSearch.asp - accessed on April 5, 2015

Table 2 - Nuclear Science Division LBNL - http://ie.lbl.gov/fission/252Cf.txt - accessed on April 5, 2015

3 - Nuclear Science Division LBNL - http://1.usa.gov/1FvmSvn - accessed on April 5, 2015