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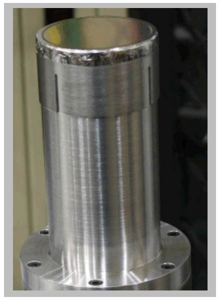

Doppelgänger Radionuclides and Mono-Energetic Electrons

Operational HP Issues at Sandia's RITS-6 Accelerator

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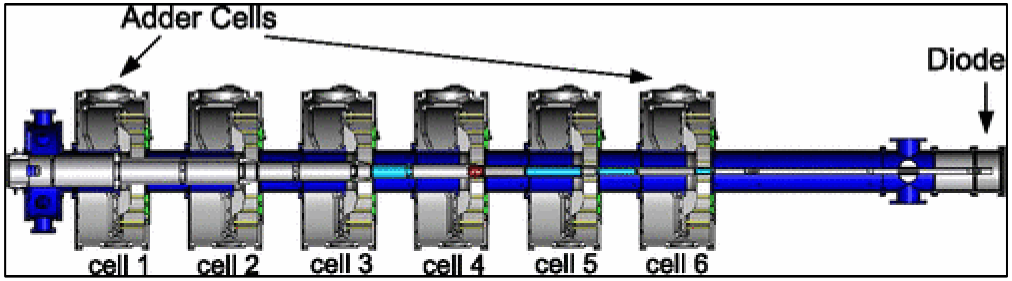
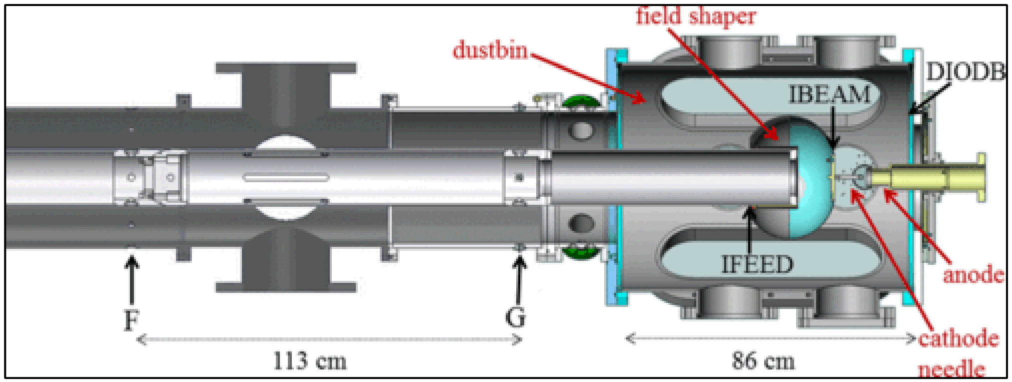
Operational HP Issues at RITS-6

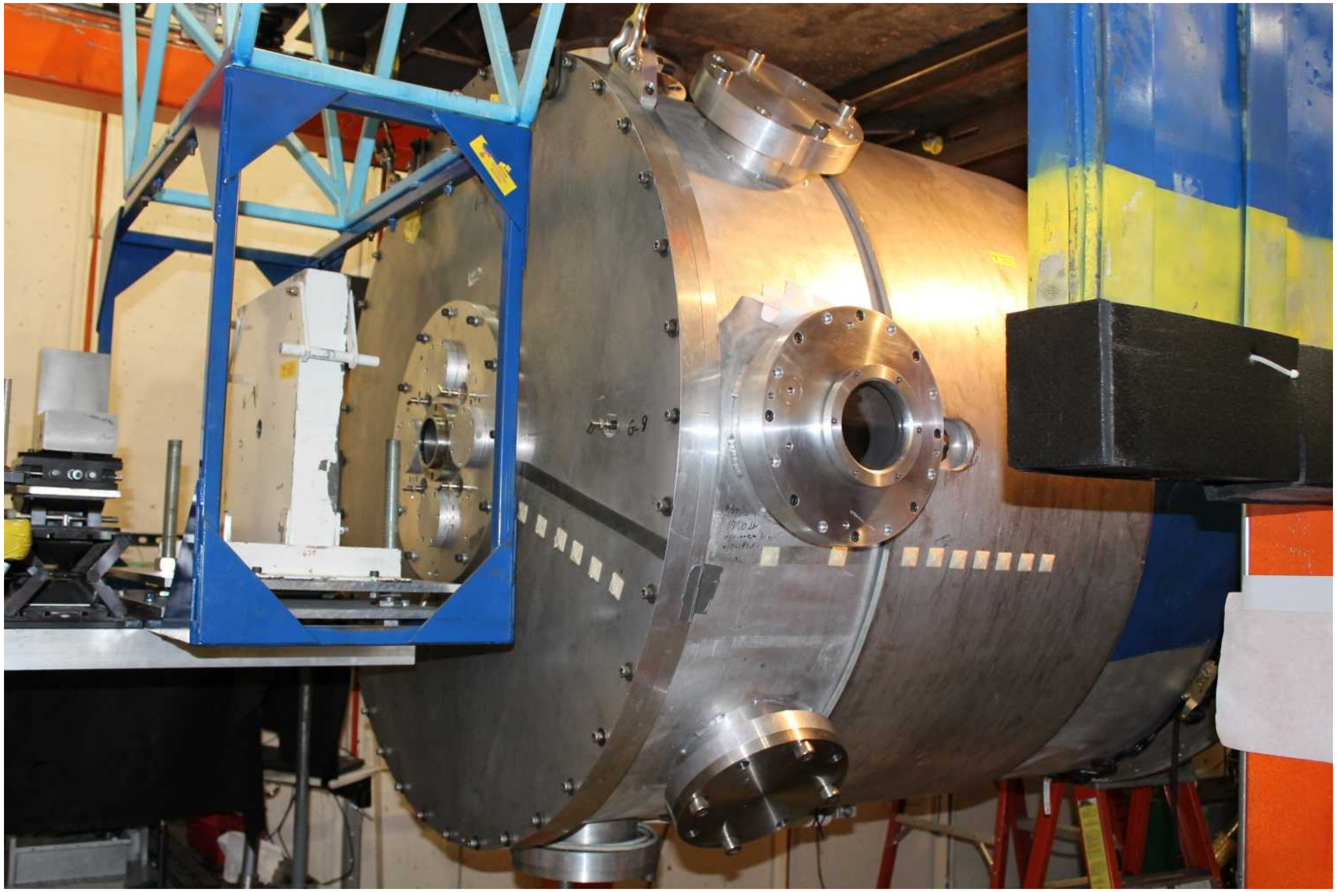
- Overview of the RITS-6 accelerator
- Shot turn-around
- Doppelgänger radionuclides
- Counting mono-energetic electrons
- Operational advantage to RITS-6





Part 1: Introduction

OVERVIEW OF THE RITS-6 ACCELERATOR



Exterior of “Dust Bin” Vacuum Chamber

Collimator and experimental target to right

Overview of the RITS-6 Accelerator

- Radiographic Integrated Test Stand
  - Second generation of RITS
    - RITS-3 had three accelerating cavities (cells)
    - RITS-6 has six
  - First operation of RITS-6 in 2005
- Usually just called RITS
- Located in Sandia's Tech Area IV
- Operated by Sandia National Laboratories (SNL) and National Security Technologies, LLC (NSTec) on behalf of DOE/NNSA

Overview of the RITS-6 Accelerator

- Experimental purpose
  - Test bed for pulsed-power radiography technology
    - Understanding IVA accelerators
      - Inductive Voltage Adder
    - Experimental diodes
      - architecture and materials
  - High power pulsed radiography
    - 45 ns pulse width
    - < 1 cm x-ray spot size
    - High spatial and temporal resolution
      - In dense and dynamic materials

Overview of the RITS-6 Accelerator

- Physical description
  - Linear electron accelerator
    - One Marx generator with 36 capacitors
    - Two intermediate storage capacitors and laser-triggered gas switches
    - Six pulse forming lines
    - Six voltage adder cells
    - High or low-impedance MITL power-flow surfaces
      - Magnetically Insulated Transmission Line
    - 6 – 11 MV accelerating potential

Overview of the RITS-6 Accelerator

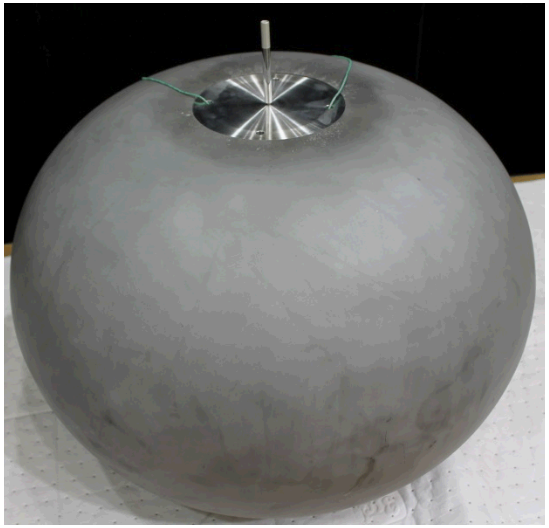
- Physical description
  - Bremsstrahlung x-ray secondary beam
    - Electron beam fired into primary target
      - Sometimes called the converter
      - Usually tantalum (Ta)
      - Sometimes tungsten (W)
    - Secondary x-ray beam irradiates the experiment
      - Also called target
    - X-ray spot size < 1 cm
    - End-point energy 6 – 11 MeV

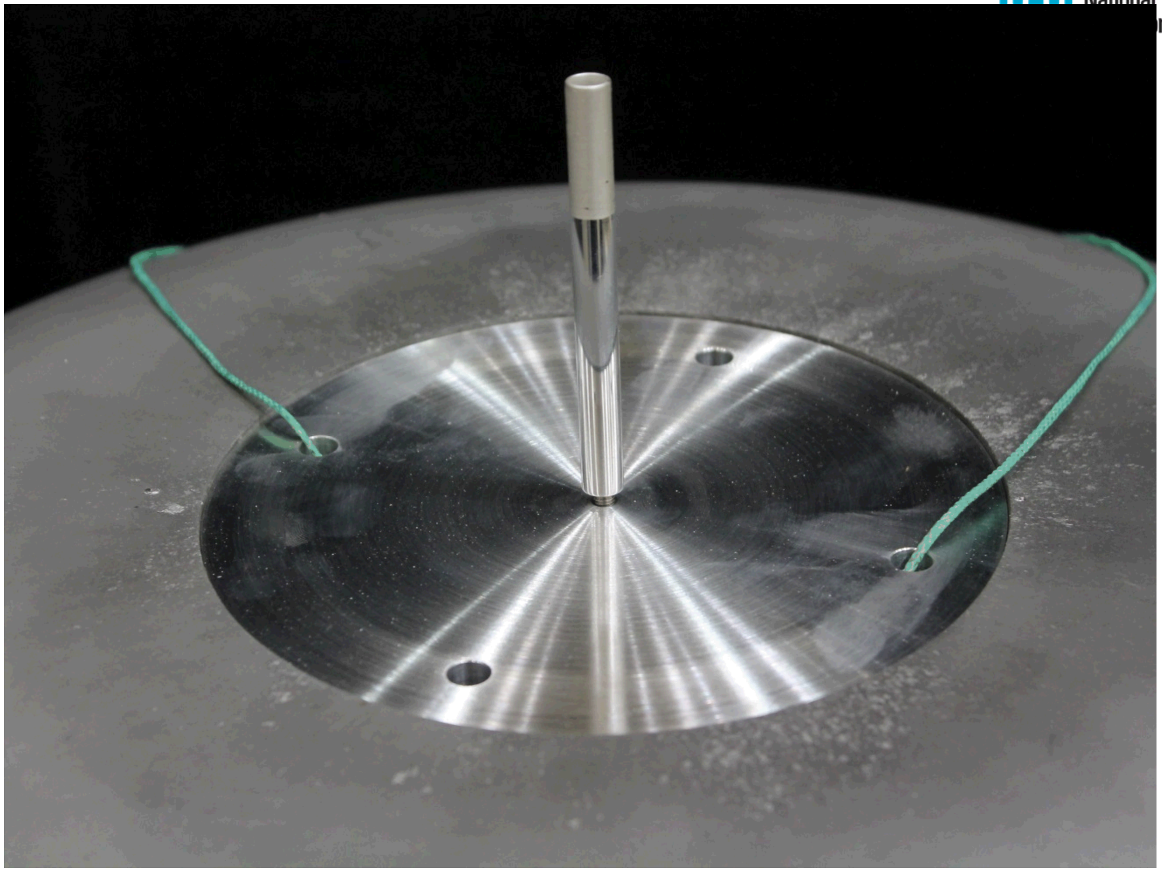
Overview of the RITS-6 Accelerator

- Diode
  - Cathode and anode separated by vacuum
    - A-K gap variable, 1—2 cm
  - Materials and architecture change with experiments
  - Region of plasma formation
    - Also solid debris and molten slag
  - Most of the activation occurs here
    - Dominant reactions
      - γ, n (photo-activation)
      - p, n (ion activation)
    - Neutron activation much less important
    - Protons are created in plasma from hydrocarbon residues present on hardware

Overview of the RITS-6 Accelerator

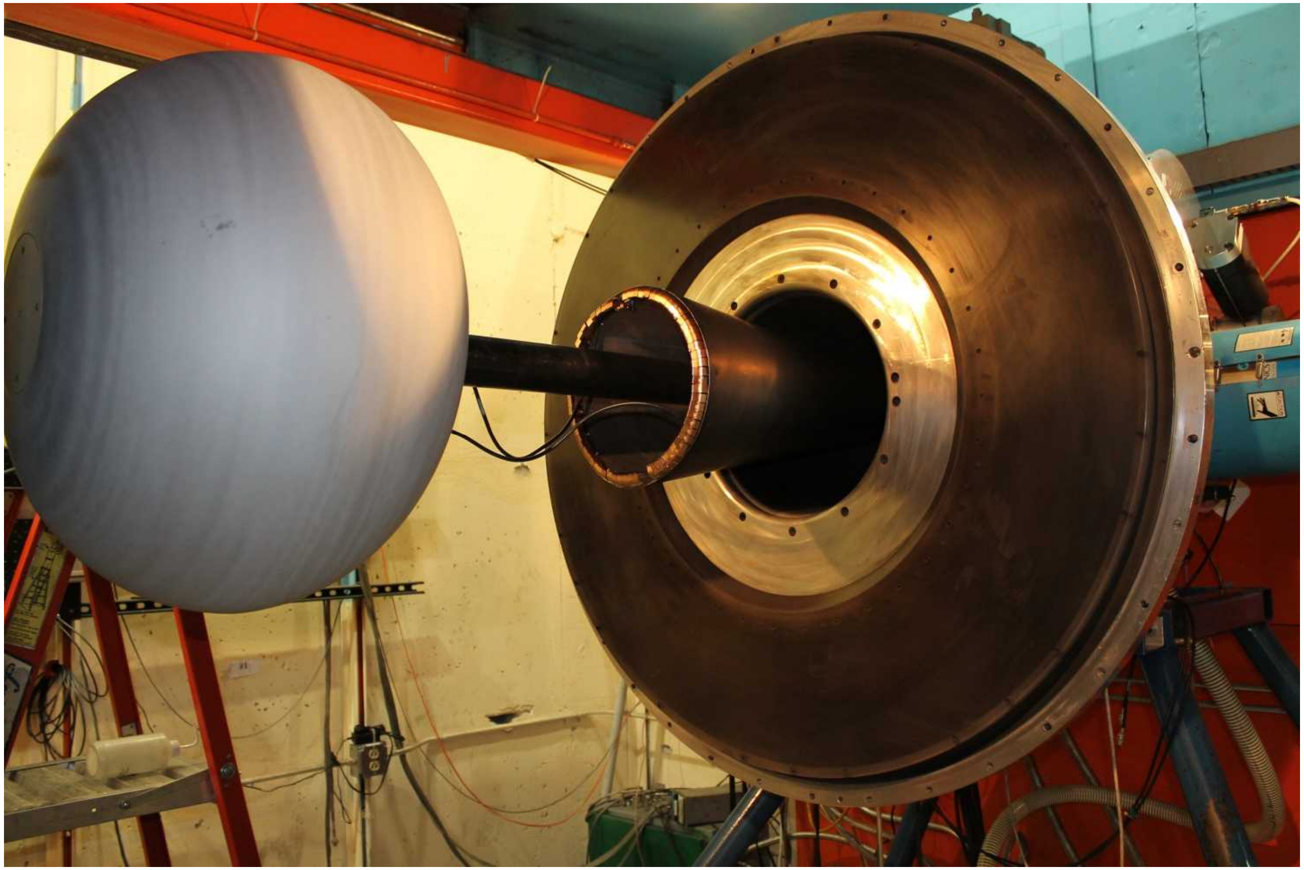
- Cathode
  - Electron current flows from MITL across a field-shaping surface called the “knob”
  - Beam emerges from a small cathode extension tube called the “needle”
  - The needle is painted with colloidal silver paint to optimize its conductivity





Silver-Painted Cathode Needle and Knob

Emergence of electron beam



Knob Mounted to MITL

Power flow and field shaping surface shown partly assembled

Cathode needle not shown

Overview of the RITS-6 Accelerator

- Anode
  - Typical converter target: 1 mm-thick tantalum (Ta)
    - Occasional substitution of tungsten (W)
  - Various beam stops and collimators

Anode

Generation of x-ray beam via bremsstrahlung on tantalum converter target

Anode Mounted to Dust Bin

Origin of x-ray beam

Part 2: Clearing the machine for re-entry

SHOT TURN-AROUND

Shot Turn-Around

- Usual shot rate is about once per day
- RITS is capable of two or more shots per day
  - Multiple daily shots would aid mission
- Some hardware must be replaced for each shot
  - Some hardware is refurbished, but multiple spares exist
  - Turn-around is not limited by refurbishment
- Induced radioactivity (activation) renders the hardware temporarily radioactive

Shot Turn-Around

- Workers enter vacuum chamber "dust bin" to retrieve and replace hardware
  - But only after contamination measurements fall below Contamination Area (CA) levels
    - Work restriction <CA imposed by the line organization
    - Radiation Protection is willing to support work in CA
    - Line personnel believe the time gained would not be worth the additional controls and slowness necessary for contamination work

Dust Bin

Open for characterization and rebuilding

Shot Turn-Around

- Worker entry allowed when:
  - β-γ < 1 kdpm/100 cm^2 removable
    - Also β-γ < 5 kdpm/100 cm^2 total
    - And dose rates < 5 mR/hr
    - Removable contamination always dominates at RITS
  - First entry by RCT ~15 minutes post-shot to assess conditions
    - Removable contamination swipes usually counted on pancake G-M (PGM) shielded detector

Shot Turn-Around

- Typical post-shot conditions
  - Conditions vary widely with machine configuration
    - Up to 100 kdpm/100 cm^2
    - 50 kdpm/100 cm^2 typical
  - Wait time for decay
    - 4 hours typical
    - Often effectively overnight

Shot Turn-Around

- Compound decay of multiple activation products
  - Early decay is rapid
    - $t_{½}$ minutes
  - Limiting decay
    - $t_{½}$ several hours

Shot Turn-Around

- What is the limiting radionuclide?
  - Early analysis identified ^{180}Ta
- ^{180}Ta
  - $t_{½}$ 8.15 hr
  - Photo-activation of tantalum
    - 99.988% of tantalum is stable ^{181}Ta
      - Remainder is ^{180m}Ta
        - » Warning:
          - » Technical references are inconsistent with each other about isomer designations
        - » $t_{½} > 1.2e15$ yr
        - » Only naturally-occurring excited-state isomer
        - » Rarest primordial isotope of any element having stable isotopes

$$^{181}Ta + \gamma \rightarrow {}^{180}Ta + n$$

Shot Turn-Around

- ^{180}Ta
  - Primary gamma 93.4 keV, 4.5% intensity
    - Confirmed present at RITS by numerous gamma-specs
  - Principal decay mode electron capture (ε) 86% → ^{180}W
    - Emits no particles directly
    - Not directly countable by PGM
      - Part 4 discusses this problem further
  - Secondary decay mode beta (β^-) 14% → ^{180}Hf
    - Mean beta energy 210 keV
    - Resembles ^{36}Cl on energy calibration curves
      - ^{36}Cl has 15% counting efficiency on PGM probe
    - Overall efficiency (14% x 15%) = 2%

Shot Turn-Around

- Ta-180
  - 2% counting efficiency
  - To see < 1 kdpm/100 cm^2
    - Must see < 20 cpm net
    - Against background of 30 – 60 cpm
  - We'll return to the low counting efficiency in part 4
- But first, we must solve a mystery!

Part 3: Induced Radioactivity at the RITS-6 Accelerator

DOPPELGÄNGER RADIONUCLIDES

The Mystery

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Bright Idea

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- Let's remove the tantalum!
  - Most x-ray devices use tungsten
  - RITS prefers tantalum for its material properties
    - But tungsten performs similarly as a converter
- Without the ^{180}Ta, turn-around should be as fast as operations can physically manage

Fizzled Idea

- Several shots were conducted using tungsten converters
- No tantalum in the chamber
- Result?
  - Still got same contamination levels
  - Still got same decay times
  - Gamma-specs still showed ^{180}Ta

What's Going On?

- Can you make ^{180}Ta on tungsten?
  - Short answer: not on RITS
- Are we activating residual tantalum in the chamber?
  - Short answer: not enough to explain persistent levels
- Have we misidentified the ^{180}Ta?
  - Maybe we should call the sleuths at RP Sample Diagnostics Lab…

Doppelgänger Radionuclides

Doppelgänger Radionuclides

- ^{180}Ta has an evil twin in ^{107}Cd

Doppelgänger Radionuclides

- Remember the silver-painted cathode needle?
- The reverse current at RITS is dominated by protons
- Protons strike the silver-tipped needle and activate it
  - Silver is 51.8% ^{107}Ag

$$^{107}Ag + p \rightarrow {}^{107}Cd + n$$

$$^{107}Cd \rightarrow (\varepsilon)\ {}^{107m}Ag \rightarrow (IT)\ {}^{107}Ag$$

  - ^{107m}Ag $t_{1/2}$ 44.2 sec

Doppelgänger Radionuclides

^{180}Ta

- $t_{½}$ 8.15 hr
- Primary gamma 93.4 keV
  - 4.5% intensity
- Other gammas too weak to measure at usual sample activities and count times

^{107}Cd

- $t_{½}$ 6.50 hr
- Primary gamma 93.1 keV
  - 4.7% intensity
- Other gammas too weak to measure at usual sample activities and count times

Doppelgänger Radionuclides

- ^{180}Ta and ^{107}Cd are indistinguishable in the field
  - Even with careful measurements
- They are also indistinguishable in the lab without extra effort
  - At usual low activities
- Enter RPSD…


F1
Help
F2

Part 4: Characterizing Instrument Response to RITS-6 Contaminants

COUNTING MONO-ENERGETIC ELECTRONS

^{107}Cd: Friend or Foe?

- Having learned that we have ^{107}Cd contamination – in addition to, or instead of, ^{180}Ta – is that good news or bad?
  - How does the counting efficiency for ^{107}Cd compare to that for ^{180}Ta?
  - What proportions of the two contaminants are present?

^{107}Cd Counting Efficiency

- Recall that we count ^{180}Ta with 2% efficiency
  - 14% β^- branch x 15% instrument eff. = 2%
- Problem: determine counting efficiency for ^{107}Cd
- We tried and rejected two solutions before settling on a third

^{107}Cd Counting Efficiency

- **Approach 1**: Consider only **beta** emissions
  - i.e., calculate for ^{107}Cd the same way as we have done for ^{180}Ta
- ^{107}Cd decays to ^{107m}Ag by two branches
  - Electron capture (ε) 99.8%
  - Positron emission (β^+) 0.2%
- ^{107m}Ag decays back to stable ^{107}Ag by isomeric transition (IT) 100%
  - No betas from this decay

^{107}Cd Counting Efficiency

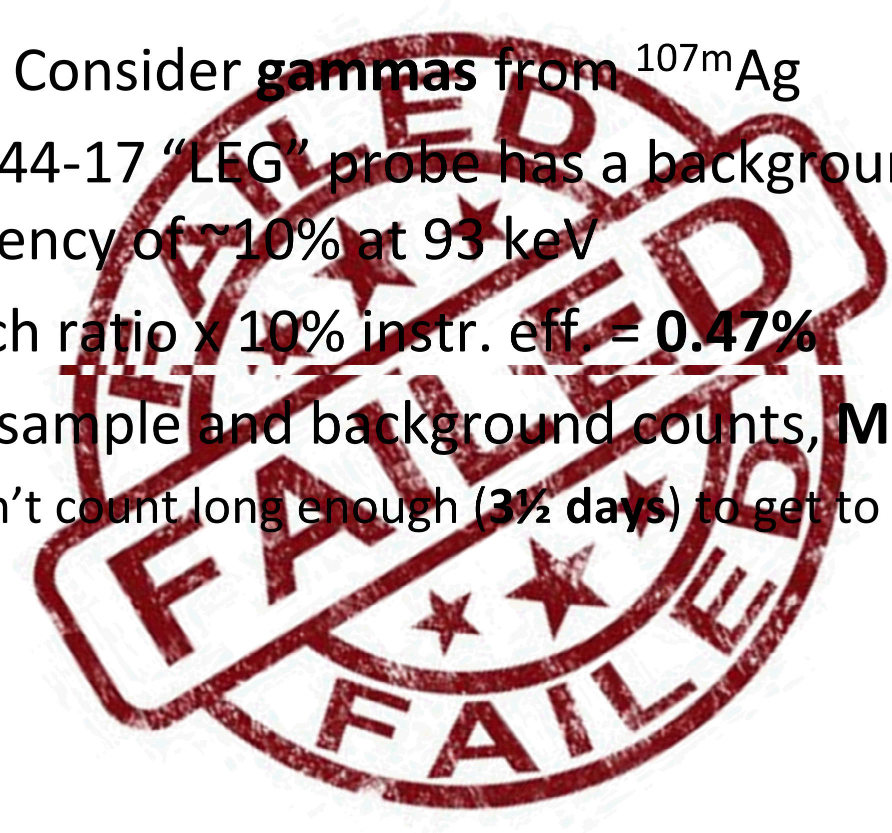
- **Approach 1**: Consider only **beta** emissions
- ^{107}Cd resembles ^{137}Cs on our calibration curve
  - ^{137}Cs efficiency 14%
- 0.2% β branch ratio x 14% instr. eff. = **0.03%**
- With 1 min. sample and background counts, **MDA > 135 kdpm** (with bkgd = 60 cpm)
  - We can’t count long enough (**25 days**) to get to 1 kdpm

^{107}Cd Counting Efficiency

- **Approach 2**: Consider **gammas** from ^{107m}Ag
- ^{107m}Ag decays back to stable ^{107}Ag by isomeric transition (IT) 100%
- Isomeric Transition is the decay mode responsible for gamma rays
  - 93.1 keV with 4.7% yield in the case of ^{107m}Ag
- Perhaps we can count the ^{107}Cd with a gamma probe?

^{107}Cd Counting Efficiency

- **Approach 2**: Consider **gammas** from ^{107m}Ag
- The Ludlum 44-17 “LEG” probe has a background of ~2 kcpm and an efficiency of ~10% at 93 keV
- 4.7% γ branch ratio x 10% instr. eff. = **0.47%**
- With 1 min. sample and background counts, **MDA > 45 kdpm**
  - We still can’t count long enough (**3½ days**) to get to 1 kdpm



Mystery #2

Mystery #2

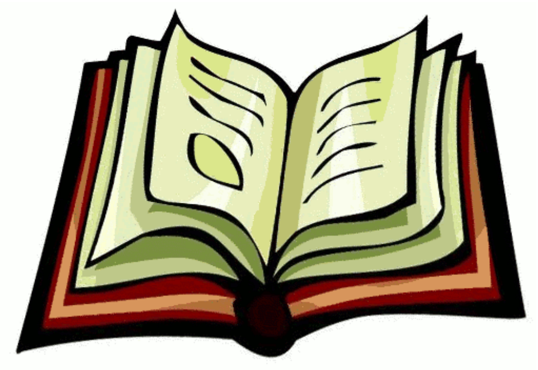
- It seems like we can't count ^{107}Cd without first dying of old age
- But two things kept nagging at us
  - Common sense
  - Physics

Mystery #2

- **Common sense** reality check:
- If the counting efficiency is really so poor for ^{107}Cd and its progeny, how come are we seeing **so much response** on the instrument?
  - Consider the implication of the “betas only” approach:
  - If we measure 100 kdpm @ 2% (assuming ^{180}Ta) but we’re really counting ^{107}Cd, that implies we have nearly **7 Mdpm** ^{107}Cd at 0.03%
    - We’re pretty sure that’s not the case

Mystery #2

- **Physics** reality check:
- If ^{107m}Ag decays 100% by Isomeric Transition, how come it emits a gamma **only 4.7%** of the time?
  - Isn't IT the same thing as gamma decay?
  - What becomes of the **other 95.3%**?
- If I remember anything from college, conservation of energy would be high on that list
- Time to crack the old textbooks!



Isomeric Transition

- Although we associate IT with gamma emission, there is another IT mode
- **Internal Conversion (IC)** competes with gamma emission in some nuclides
  - Gamma emission is a sub-mode of IT
  - IC is a sub-mode of IT
- In **IC**, the excitation energy present in the isomer’s nucleus, instead of appearing as a gamma ray, is transferred to an orbital electron, which is ejected forcefully from the atom

Conversion Electrons

- The ejected electron is called a **conversion electron**
  - It carries an energy quantum equal to what a gamma would have carried from the same transition, less the binding energy
  - The binding energy is usually << the radiated energy
  - Some nuclides emit gammas and conversion electrons of essentially equal energy

Conversion Electrons

- How do conversion electrons differ from betas?
  - Internal conversion is a **two-body** problem
    - Daughter nucleus
    - Conversion electron
  - Contrast with beta decay, a **three-body** problem
    - Daughter nucleus
    - Beta particle
    - Neutrino
  - In a two-body problem, conservation of energy and momentum gives a **single** solution
    - In a three-body problem, a **range** of solutions exist
  - Where beta decay yields a range of energies, IC yields an electron at a definite (discrete) energy

Auger Electrons

- In the same way **IC** competes with **gamma** emission for excited **nuclei…**
- The **Auger Effect** competes with **x-ray** emission for excited **atoms**
- Excitation energy in the form of an electron-shell vacancy can be emitted, not as an x-ray, but as an **Auger electron**
- The Auger Effect is also a two-body problem
  - Daughter nucleus
  - Auger electron
- Thus Auger electrons are emitted, like conversion electrons, at definite discrete energies

Mono-Energetic Electrons

- Conversion and Auger electrons are mono-energetic
  - They are called electrons, not betas
- Most nuclei have several varieties of emitted electrons, but each variety corresponds to a specific nuclear or atomic transition
  - Each is emitted at a discrete energy

Counting Mono-Energetic Electrons

- A particle detector does not know the origin of the particle it counts
  - It doesn't care if it was a beta or an electron
- Most Auger electrons, and some conversion electrons, have too little energy to be detected by contamination probes
- But the ones that are sufficiently energetic get counted just like betas of comparable energy

Counting Mono-Energetic Electrons

- Ignoring electrons can be a serious mistake
  - Yet HPs rarely discuss them
- In the case of ^{107}Cd, it makes a difference of **hundreds-fold** in the counting efficiency
  - The "beta only" view gives an efficiency of 0.03%
  - Including the electrons gives an efficiency of 10%
- In the case of ^{180}Ta, it **doubles** the efficiency
  - 4% vs. 2%

Efficiency vs. Energy

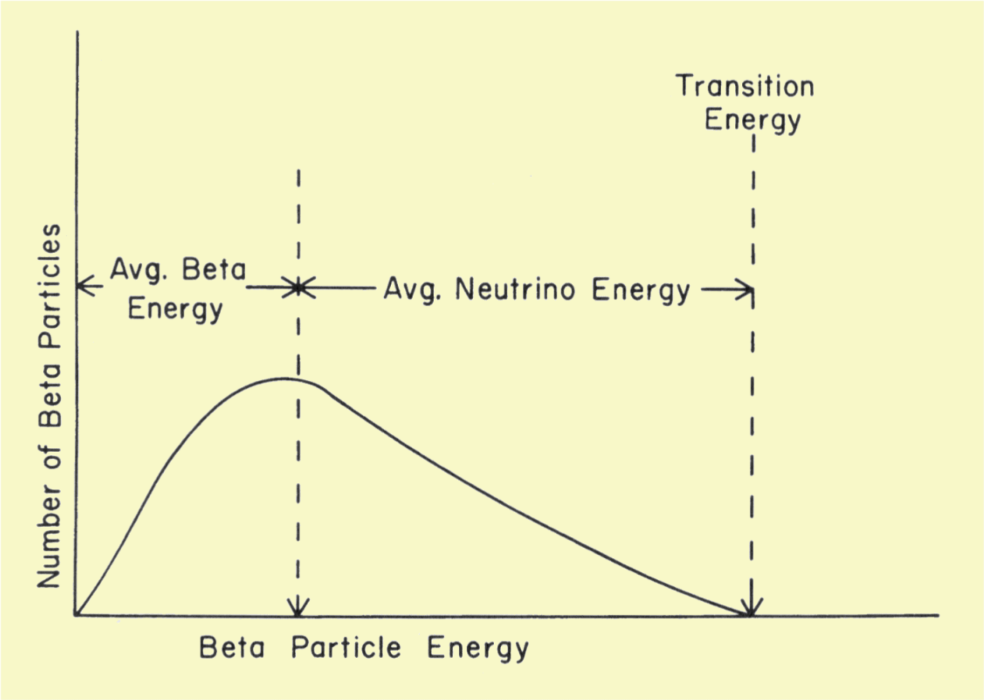
- A particle detector cares only about the amount of energy carried by the particle when it arrives at the detector
  - Not its origin or what we call it
  - In some detectors, it matters only that it got into the sensitive volume
    - The energy affects whether it makes it in
- In contamination probes, counting efficiency generally rises with energy
  - Sharply so at low energies

Efficiency Curves

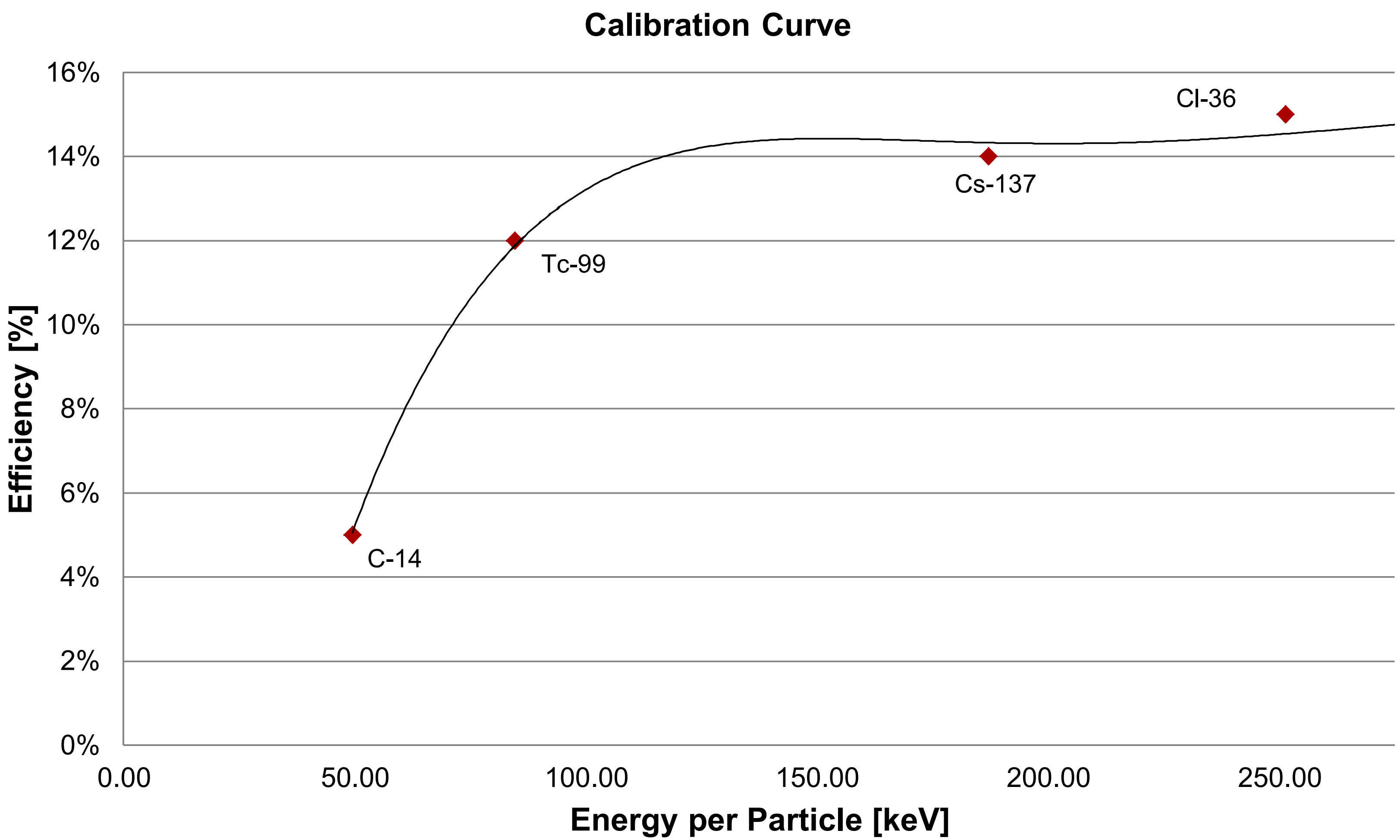
- Efficiency calibration curves are measured with known **beta** sources
- For other **beta** emitters, we interpolate between the calibrated energies
  - Usually by reference to maximum energies
    - It is probably more accurate to interpolate by reference to average energies

Efficiency Curves

- For **electron** emitters, we interpolate as before, but with reference to beta average energies



Efficiency Curve for a PGM



Energy Comparisons

- The detector cares, in terms of its efficiency, about the energy of the countable particles
- We calculate efficiency for a nuclide in three steps
  - Screen out uncountable and low-abundance particles
  - Compare by interpolation on calibration curve, for each countable particle
    - β: average energy per particle
    - Conversion electron: particle energy
    - Auger electron: particle energy
  - Multiply counting efficiency times particle abundance
  - Sum the products for total efficiency for that nuclide

Particle Screening

- Screen uncountable and low-abundance particles
  - For the PGM, consider only those particles whose
    - Energy, $E > 40$ keV
      - This reflects the weakest detectable particle on the PGM
    - Abundance $A > 0.01\%$
      - This is an arbitrary value

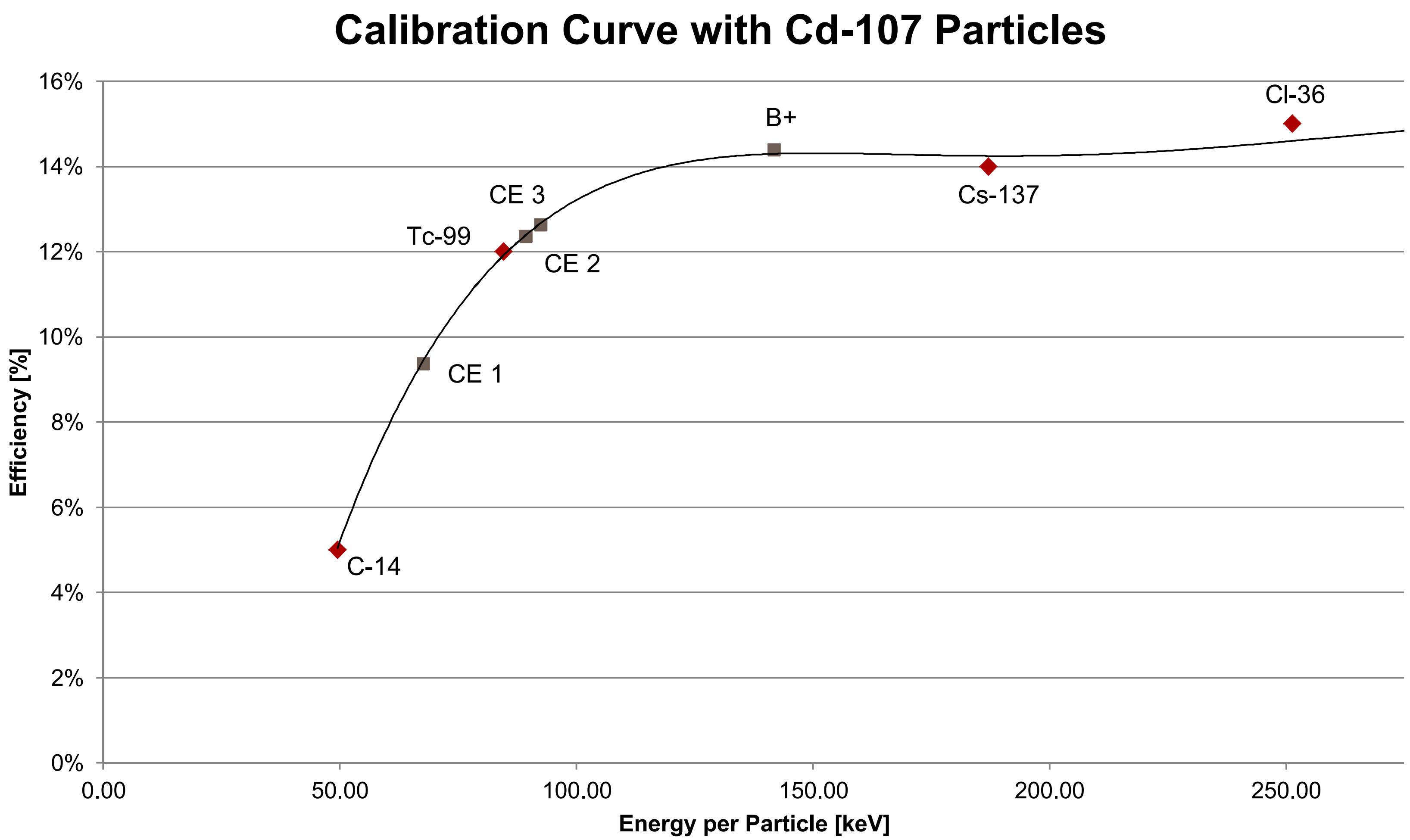
Total Efficiency

- Calculate the total efficiency for all countable particles

$$Eff = \sum_i Eff_i A_i$$

- Where Eff_i is the particle-specific efficiency
- And A_i is the particle-specific abundance

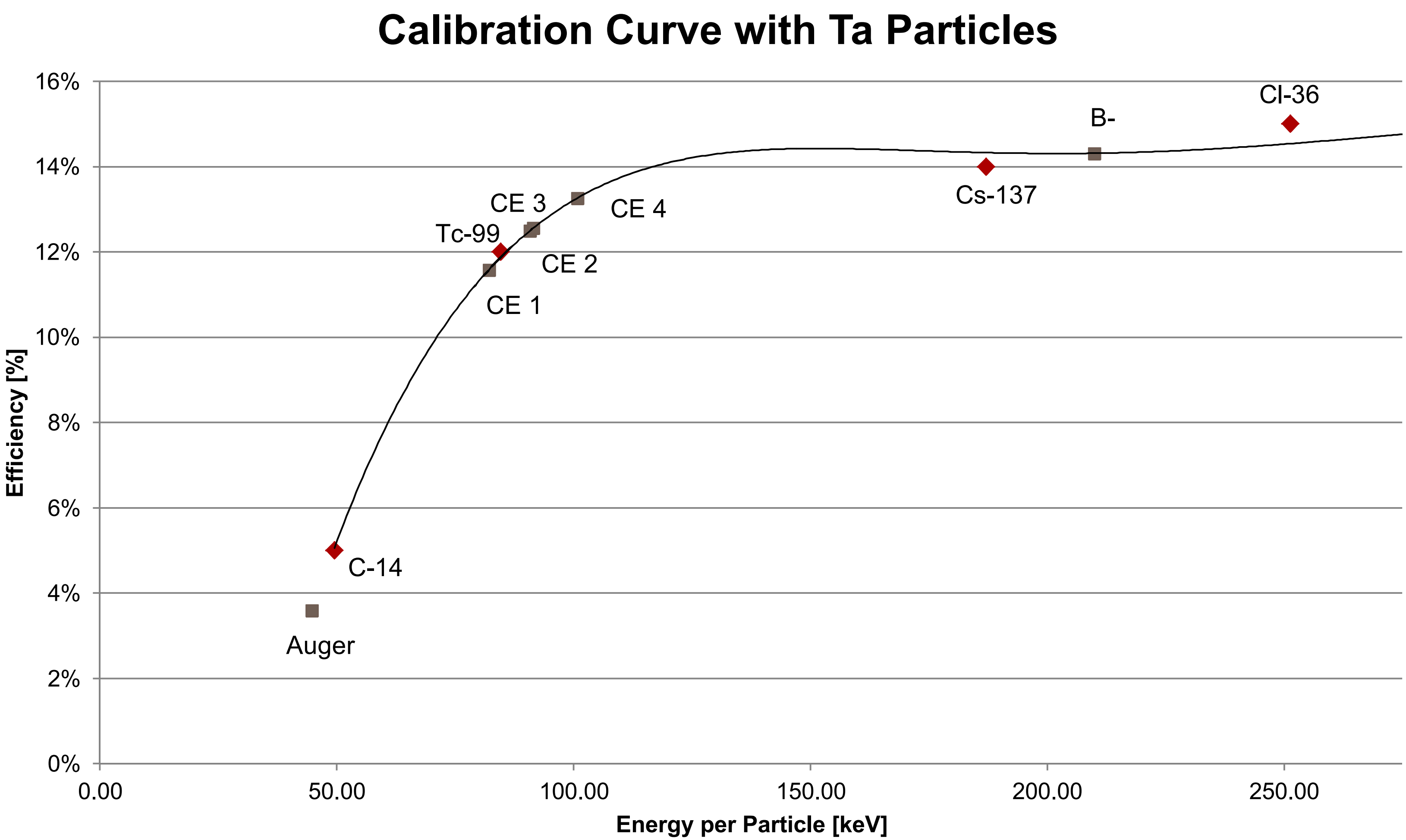
Calculation for ^{107}Cd



Calculation for ^{107}Cd

^{107}Cd				
	Energy [keV]	Abundance	Instrument Efficiency (Interpolated)	Particle Efficiency
CE 1	67.61	0.44	9.38%	4.13%
CE 2	89.318	0.41	12.36%	5.07%
CE 3	92.407	0.084	12.64%	1.06%
β^+	141.8	0.002	14.40%	0.03%
			Total Efficiency	**10.28%**

Calculation for ^{180}Ta



Calculation for ^{180}Ta

^{180}Ta				
	Energy	Abundance	Instrument Efficiency (Interpolated)	Particle Efficiency
Auger	44.8 keV	0.036	3.59%	0.13%
CE 1	82.13 keV	0.12257	11.58%	1.42%
CE 2	90.8 keV	0.030463	12.50%	0.38%
CE 3	91.5 keV	0.016	12.56%	0.20%
CE 4	100.78 keV	0.004	13.26%	0.05%
β^-	210 keV	0.136	14.31%	1.95%
			Total Efficiency	**4.13%**

Part 5: Why this matters

OPERATIONAL ADVANTAGE TO RITS-6

Proportions between ^{107}Cd and ^{180}Ta

- We don’t know yet what proportion of the two contaminants we’re producing
- We hope to get an electron-spectroscopy analysis soon
- Several indications suggest that cadmium dominates
- For now, we’ll conservatively assume a 50/50 split

Advantage to RITS

- This yields a **7%** counting efficiency
  - Compared to the old 2%
- This **3½ times gain** in efficiency means waiting 1.8 fewer half-lives
- This is a gain of **>10 hours** turn-around (ideally)
  - Not all that gain will be realized
  - Should effectively eliminate the long-lived waiting

Questions?

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