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Title: MCNPX Simulation of a Passive Gamma System to be Used in a Spent Fuel Plutonium Assay Strategy

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MCNPX Simulation of a Passive Gamma System to be Used in a Spent Fuel Plutonium Assay Strategy

Jack Douglas Galloway

Michael Lorne Fensin

Abstract

MCNPX has been used to simulate the response of a passive gamma system used in the assay of spent nuclear fuel. This talk, presented for the Next Generation Safeguards Initiative (NGSI) review committee, first addresses source set-up and transport simulation challenges and methods, and then examines how signal emission ratios from fission products can be used to quantify attributes of spent nuclear fuel. Specifically examined in this presentation are $^{134}Cs/^{137}Cs$ as well as $^{154}Eu/^{137}Cs$ ratios from specific spent nuclear fuel assembly burnups, initial enrichments and cooling times. Results show that ratios can be used to effectively ascertain state-point information.

Passive Gamma
Presented by Jack Galloway

Paper prepared by:
Michael L. Fensin, Stephen J. Tobin, Martyn T. Swinhoe, Howard O. Menlove, Will Koehler, Nathan P. Sandoval, Sang Lee, Vladimir Mozin, Josh Richard, Melissa Schear, Jianwei Hu, Jeremy Conlin

Passive Gamma

- **Spent fuel assemblies (actinides and fission products), among numerous radioactive releases, also emit:**
  - Spontaneous, delayed and (α,n) neutrons
  - Gamma rays during the decay of fission product
- **These assemblies are termed as a "source" of neutrons and gammas for further transport calculations**
  - Active interrogation: exposing the fuel to some external source of particles and analyzing the resultant spectra
    - Provide a snapshot in time
  - Passive interrogation: particles and interactions that occur as a result of radioactive decay of the assembly and analyzing the resultant spectra
    - Provide time integrated effects up to snapshot
- **Passive gamma (PG) is the detection and interpretation of gamma lines emitted spent nuclear fuel (SNF)**

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SNF Source Characteristics

- **SNF gamma emission signal is dependent upon the spatial and temporal behavior of the radioactive constituents within the fuel**
  - This behavior is dictated by fission yield, capture cross section, activity and branching ratio
  - Energy emission affects attenuation
    - ^{134}Cs (795.8 keV), ^{137}Cs (661.7 keV), ^{154}Eu (1274.4 keV)
- **Leverage the N-4 SNF library**
  - M. L. Fensin, et al., "A Monte Carlo Linked Depletion Spent Fuel Library for Assessing Varied Nondestructive Assay Techniques for Nuclear Safeguards," ANFM IV, (2009)

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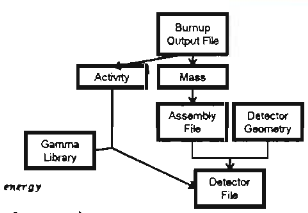
PG Source Generation

- The count time is relatively short compared to the prior decay time leading to the count (S.S. picture)
- Only the most significant lines are important
  - FPs from SNF that have large signal to background ratio



$$if\ \frac{A_{i,r} \cdot BR_{j,i,r}}{A_{MAX,r} \cdot BR_{MAX,i,r}} > \varepsilon\ then\ keep\ emission\ energy$$

$$\%contribution = \frac{\sum_{i=0}^{N}\left(A_{i,r} \cdot \sum_{j=0}^{G} BR_{j,i,r}\right)}{\sum_{r=1}^{N}\sum_{i=0}^{N}\left(A_{i,r} \cdot \sum_{j=0}^{G} BR_{j,i,r}\right)}$$

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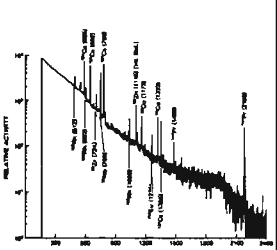
PG Simulations

- **Initial runs had 1E-4 contribution threshold**

$$if\ \frac{A_{i,r} \cdot BR_{j,i,r}}{A_{MAX,r} \cdot BR_{MAX,i,r}} > \varepsilon\ then\ keep\ emission\ energy$$

  - Referenced to maximum peak (^{137}Cs peak for this library)
  - Due to computational limitations at the time
  - Cut-off chosen from adjacent plot



* D. Reily, N. Ensslin H. Smith, and S. Kreiner, *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550, Washington, DC (1991)

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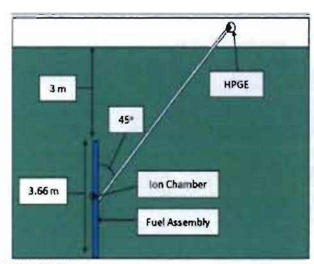
PG Simulations

- **Passive gamma counting system**
  - HPGE detector
    - 7.5 X 7.5 cm crystal
    - 1 mm thick Al window
    - 2.54 cm lead shield
  - Ion Chamber
    - N2 7600 torr
    - 3.81 cm diameter
    - 14.96 cm length
    - 1 mm thick SS casing
  - 5.08 cm radius pipe
    - 1 mm SS thickness



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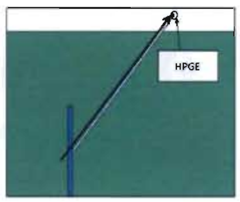
PG Simulations

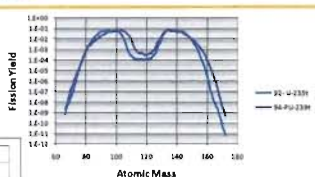


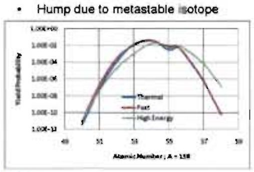
PG Simulations

- **~100 PG cases run to quantify capability of PG to detect fissile mass**
  - ~33 diversion cases
  - 64 non-diversion cases
    - Unique combination of
      - Burnup: 15, 30, 45 and 60 GWd/tU
      - Initial enrichment: 2, 3, 4, and 5%
      - Cooling time: 1, 5, 20 and 60 years
- **Gamma transport dependent upon linear attenuation coefficient of transporting medium**
  - Borated H_2O used

PG Results – Fission Yields

- **For a given atomic mass:**
  - Isotopic fission yields can vary drastically
  - 6 orders of magnitude from Z=51 to Z=53
  - Hump due to metastable isotope

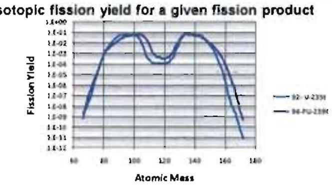




PG Results – Fission Yields

- **Drastic difference in isotopic fission yield for a given fission product**

Mass yields for ^{235}U and ^{239}Pu with respect to A=154 are quite similar, yet isotopic fission yields for ^{154}Eu are drastically different



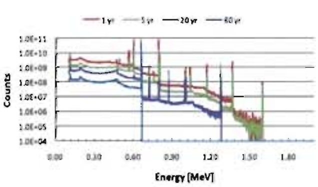
	[illegible]	[illegible]	[illegible]
[illegible]	3.86E-08	3.35E-06	1.16E-07
[illegible]	6.00E-04	5.97E-03	9.24E-04
[illegible]	9.70E-10	1.40E-03	1.54E-09

* M. B. Chadwick, et al. "ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology", Nuclear Data Sheets, 107, pp. 2931-3060 (2006)

PG Results – Gamma Spectrum

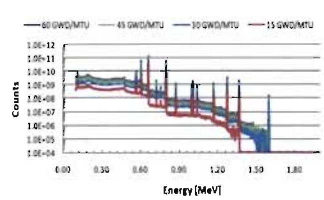
- **Activity at 30 GWd/MTU, 5% enriched**
  - Function of cooling time

- Only produce most important immediate daughter lines from Tier 2 fission product masses
- Cs-137 is the only available line above background at 60 yrs
  - All other peaks are less than 1E-4 of ^{137}Cs



PG Results – Gamma Spectrum

- **Activity at 5% enriched, 5 year cooling time**
  - Function of burnup
  - ^{154}Eu signal (1.597) less than 1E-4 of ^{137}Cs at 15 GWd/MTU, line not included



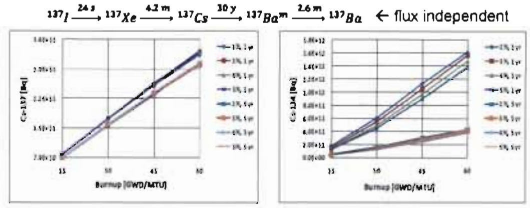
PG Results - Cesium

- **Cesium ratios**
  - ^{137}Cs and ^{134}Cs are dominant contributors
  - Ratio have been examined for quantifying bumup
    - S. T. Hsue, T. W. Crane, W. L. Talbert, J. C. Lee, "Nondestructive Assay Methods for Irradiated Nuclear Fuels," *LA-6923*, Los Alamos, NM (1978).
    - C. S. Tsao and L. K. Pan, "Reevaluation of the Burnup of Spent Fuel Pins by the Activity Ratio of $^{134}Cs/^{137}Cs$," *Application of Radiation and Isotopes*, 44, pg. 1041-1046 (1993).
    - C. Willman, A. Hakansson, O. Osifo, A. Backlin, S. J. Svard, "A Nondestructive Method for Discriminating MOX Fuel from LEU Fuel for Safeguards Purposes," *Annuls of Nuclear Energy*, 33, pg. 766-773 (2006).
  - Entire significantly contributing ^{134}Cs signal used
    - 8 of most prominent peaks considered
      - 0.475, 0.563, 0.569, 0.605, 0.796, 0.802, 1.168, and 1.365 MeV
  - ~2 year half-life of ^{134}Cs reduces usefulness with increasing cooling time
- **Power $\propto \Sigma_f * \Phi$**
  - Inverse relationship between macroscopic fission cross-section and flux for a given power

PG Results - Cesium

Governing equation for ^{137}Cs production:

$$^{137}I \xrightarrow{24\,s} {}^{137}Xe \xrightarrow{4.2\,m} {}^{137}Cs \xrightarrow{30\,y} {}^{137}Ba^{m} \xrightarrow{2.6\,m} {}^{137}Ba$$ ← flux independent



Governing equation for ^{134}Cs production:

$$^{133}I \xrightarrow{21h} {}^{133}Xe \xrightarrow{5.27d} {}^{133}Cs(n,\gamma){}^{134}Cs \xrightarrow{2.06y} {}^{134}Ba$$ ← flux dependent

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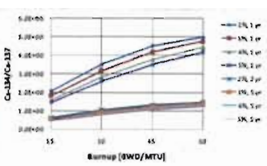
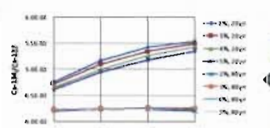
PG Results - Cesium



$^{134}Cs/^{137}Cs$ ratio 1 & 5 year cooling time



$^{134}Cs/^{137}Cs$ ratio 20 & 80 year cooling time

$^{134}Cs/^{137}Cs$ varies 16-30% as a function of IE for given BU; 58-65% as function of BU for given IE (1-5 yr cooled)

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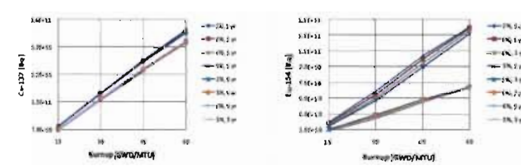
PG Results - Europium

- Governing equation for ^{137}Cs production:

$$^{137}I \xrightarrow{24\,s} {}^{137}Xe \xrightarrow{4.2\,m} {}^{137}Cs \xrightarrow{30\,y} {}^{137}Ba^{m} \xrightarrow{2.6\,m} {}^{137}Ba$$ ← flux independent



- Governing equation for ^{154}Eu production:

$$^{153}Sm \xrightarrow{46.7\,h} {}^{153}Eu(n,\gamma){}^{154}Eu \xrightarrow{8.6\,y} {}^{154}Gd$$ ← flux dependent

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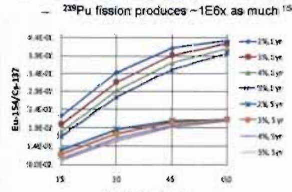
PG Results - Europium

- **Europium ratios**
  - ^{154}Eu has ~8.6 year half-life
  - ^{154}Eu production depends on fission product yield
    - ^{239}Pu fission produces ~1E6x as much ^{154}Eu as ^{235}U



$^{154}Eu/^{137}Cs$ IE variation is not as BU dependant as $^{134}Cs/^{137}Cs$ (<14%); For given IE BU signal varies 20-40%

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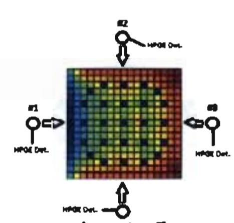
PG Next Steps

- **Spent Fuel Library #2**
  - Each pin in ½ reflected assembly burned
  - Fuel rotated in a "realistic" core shuffling sequence
    - Investigate asymmetric effects
- **Apply the same approach for PG simulations to SFL #2**
  - Detector tallies on source from 3 sides (symmetric about one mid-plane axis (HPGE #1 → HPGE #3)



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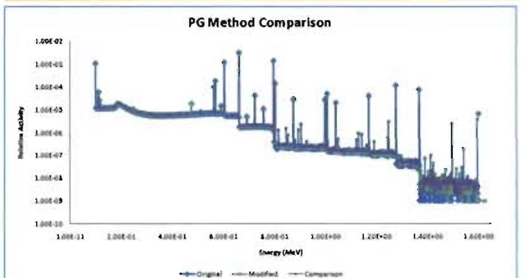
PG Next Steps

- **Increase the number of lines tracked to ensure all important lines are included**
- **Tally only gamma flux at detector surface planes**
  - These will lead to increased computational effort
  - Significant cluster upgrades since initial work afford reources for these calculations
- **Assess ability to infer**
  - Burnup
  - Initial enrichment
  - Cooling time
- **Excel macro converted to Perl script**
  - One script execution generates detector response – a "go" button!

PG Next Steps

- **Spectra comparison**
  - "Original" refers to the results from the 1E-4 threshold simulation approach initially adopted
  - "Modified" refers to the no threshold version of initial approach, including significantly more energy lines
    - Still executes source generation quite efficiently (time required for source generation << time required for MCNP execution)
- **Comparison with old method used to validate**
  - Comparison spectra determined with simulation approach developed by V. Mozin for DG research

PG Next Steps



Conclusions

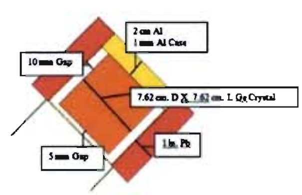
- **Use of PG for determination of burnup, initial enrichment, and cooling time ...**
  - Has been assessed for infinitely reflected assembly
    - Something on transport/source generation method
    - Something on fission yields (look at independent yield)
    - Something on ratios; flux dependence/ integrated fission dependence
- **Being evaluated for asymmetrically burned assembly**
  - Improvements due to increased computer resources ...
    - Still allows for efficient source determination
    - Includes greater number of gamma lines
- **Continuity of knowledge maintained through contact with Mike Fensin**

Done

- **Extra slides follow**

PG Simulations

- **HPGE Crystal Configuration**



Par = sp Limitations

- **MCNPX 2.7.A new passive gamma feature**
  - Generates a spontaneous photon source based on cell activity
  - Source sampling → activity*emission for a time integrated decay
  - Linked to delayed gamma capability
- **Caveats**
  - Delayed gamma capability calculates the multi-group emission spectra for both neutrons and photons regardless
  - All CDFs are not banked in MCNPX 2.7.A
    - CINDER90 is run for every source particle
    - Only certain FP decay chains are prebanked

Simple multi-cell lattice test with actinides took too long for practical use!!!

These limitations were fixed in MCNPX 2.7.D