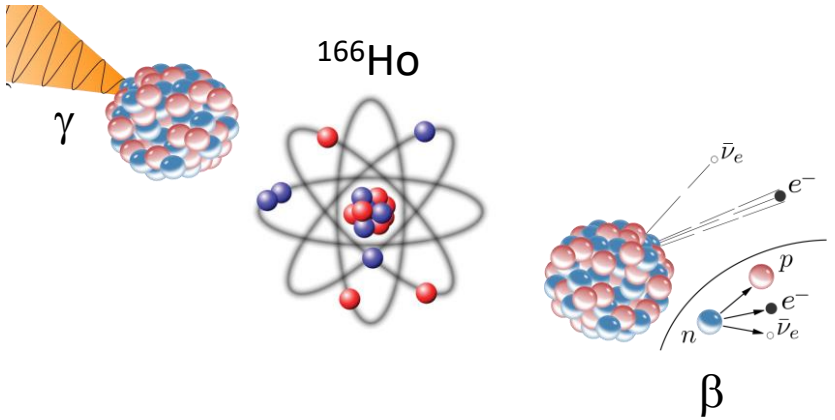


Evaluation of ^{166}Ho: a β- decay scheme updated with new measurements

Brian E. Zimmerman

Leader, Radioactivity Group
Radiation Physics Division
Physical Measurement Laboratory

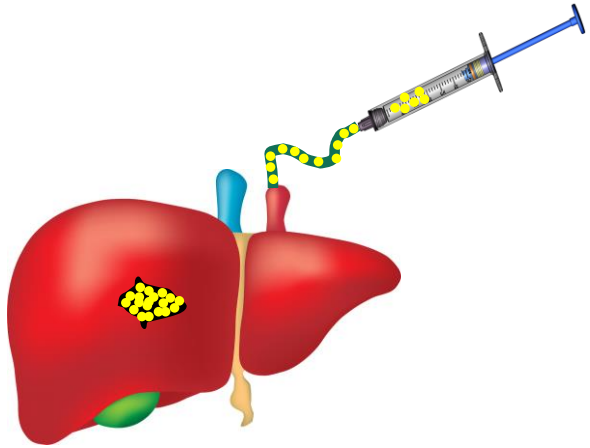
DDEP Workshop
DOSEO Saclay, CEA Saclay
21-25 October, 2024

A little about me….

- 1987-1992 Ph.D. Nuclear Chemistry, Univ. of Maryland
  - Low-energy nuclear structure: odd-odd, even-even nuclei around Z=50 shell closure
  - γ-γ, γ-e^-, time-scaled spectroscopy for level scheme development
  - Low-temperature nuclear orientation for nuclear magnetic moment measurements
  - Most work was at ORNL, Daresbury
- 1992-1995 Post-doc, Univ. Tennessee, Dept. Physics and Astronomy
  - Moment measurements via tilted foil polarization technique
  - Low-energy nuclear structure of nuclides near Z=82
  - Proton radioactivity, multi-parameter coincidence spectroscopy (mass fragments, charged particles, photons)
  - High-spin nuclear structure (GammaSphere)

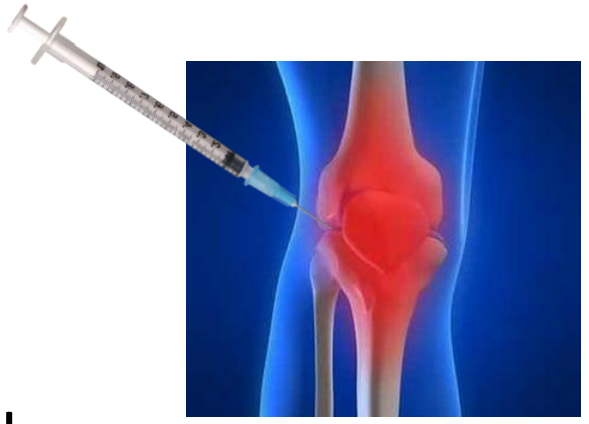
A little about me, cont'd.

- 1995-2003: Research Chemist, NIST
  - Standards for radionuclides in medicine
  - Mainly user of Nuclear Data
- 2003-2006: Medical Radiation Physicist, IAEA
  - Monte Carlo simulations
  - Dosimetry
  - Importance of nuclear data as inputs
- 2006-now: Research Chemist, NIST
  - More standards for medical nuclides
  - 2015: started involvement with DDEP – started incorporating data measurements in all standardizations
  - 2017: I-124
  - 2019: became GL of Radioactivity Group
  - 2024: Ho-166
  - 2024 (?): Pb-212
  - 2025 (?): Ra-224, Ac-225

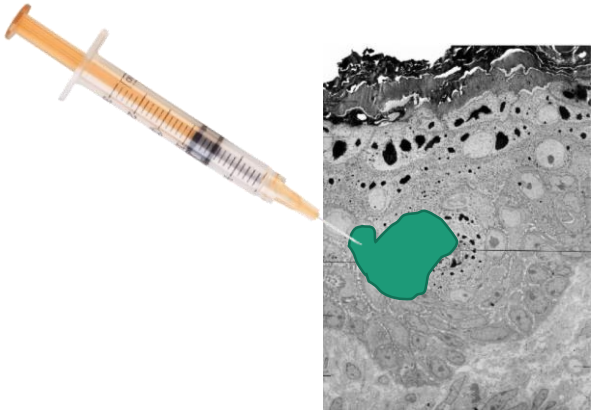


Radioactive microspheres

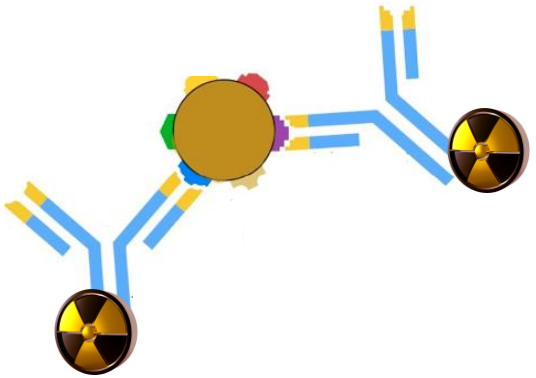
Ho-166: a nuclide with many potential medical applications because of its relatively simple chemistry



Radiation synovectomy



Intratumor injection



Radiolabeled antibodies

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Production routes

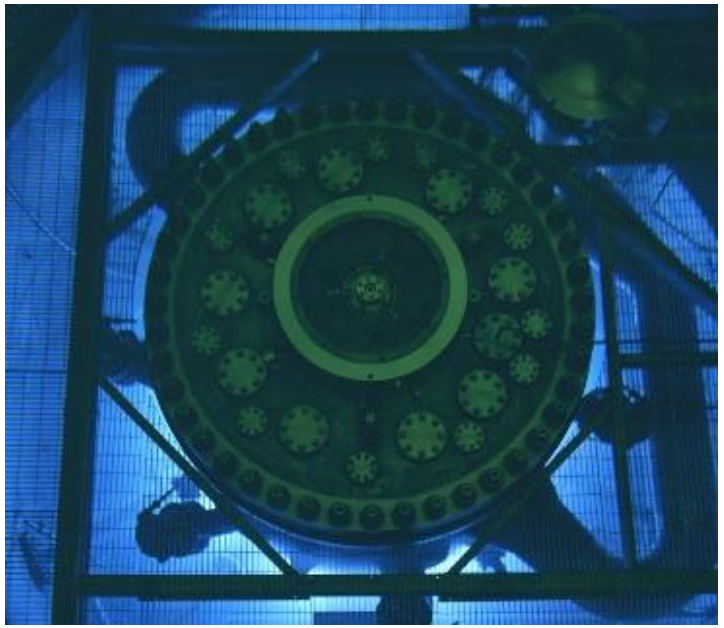
- Reactor-produced via

  $^{165}Ho(n,\gamma)^{166}Ho$

  or

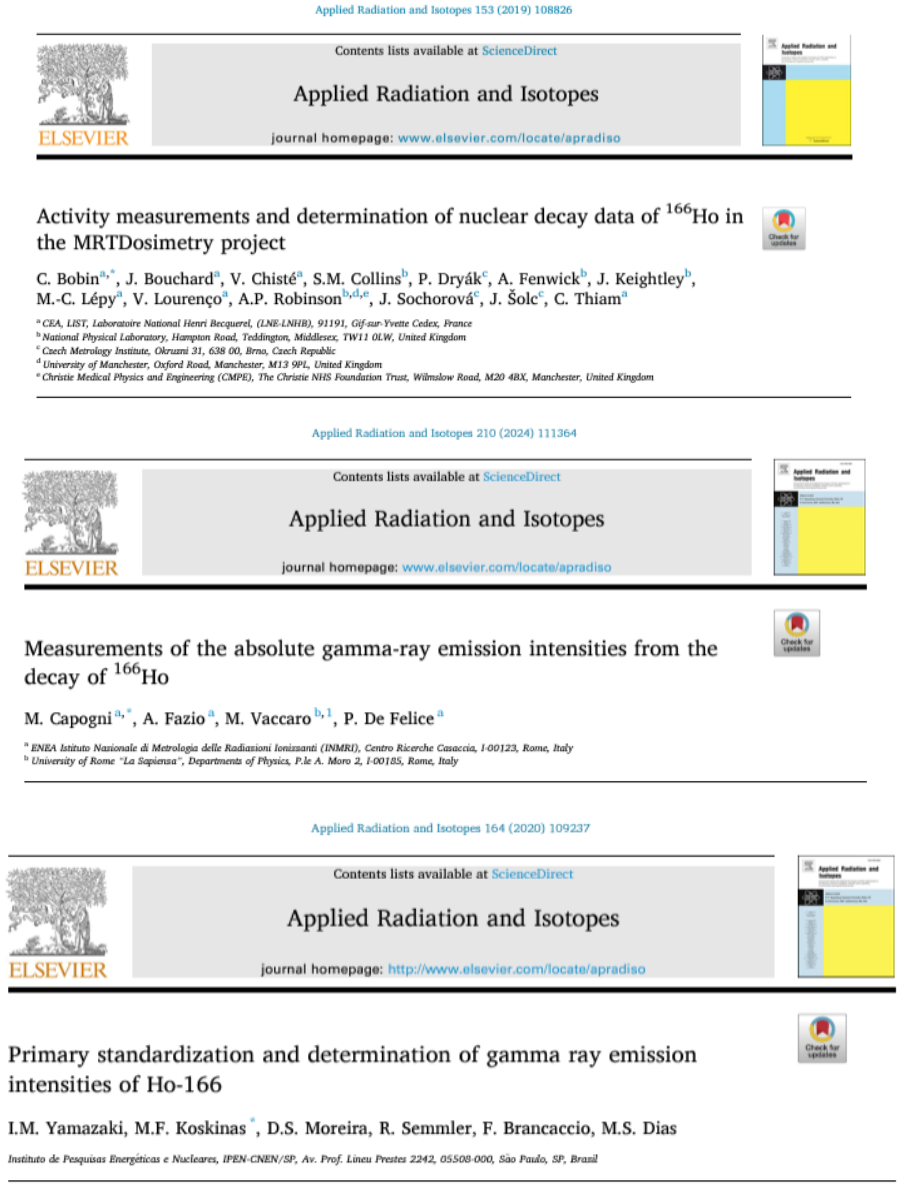
  $^{164}Dy\ (n,\gamma)^{165}Dy(n,\gamma)^{166}Dy \rightarrow {}^{166}Ho$

  reactions.
- $^{166}Dy \rightarrow {}^{166}Ho$ can function as an *in-vivo* generator
- Main impurity is long-lived (1200 a) ^{166m}Ho when produced via $^{165}Ho(n,\gamma)^{166}Ho$ reaction; no long-lived impurities from $^{164}Dy\ (n,\gamma)^{165}Dy(n,\gamma)^{166}Dy \rightarrow {}^{166}Ho$.



Previous evaluations, new data

- 2004: Previous DDEP evaluation performed by Schönfeld and Dersch (Bé et al. 2005).
- 2008: Most recent ENSDF evaluation (Baglin 2008).
- 2014: Re-assessment of previously published half-life value by NIST (Unterweger and Fitzgerald 2014).
- 2019-2024: Determinations of half-lives, X- and γ-ray emission probabilities performed by CMI, LNHB, NPL, and ENEA as part of EURAMET MRTDosimetry project (Bobin et al. 2019; Capogni et al. 2024).
- 2020: New γ-ray emission probabilities measured by IPEN (Yamazaki et al. 2020).



Applied Radiation and Isotopes 153 (2019) 108826

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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Activity measurements and determination of nuclear decay data of ^{166}Ho in the MRTDosimetry project

C. Bobin[a,*], J. Bouchard[a], V. Chisté[a], S.M. Collins[b], P. Dryák[c], A. Fenwick[b], J. Keightley[b], M.-C. Lépy[a], V. Lourenço[a], A.P. Robinson[b,d,e], J. Sochorová[c], J. Šolc[c], C. Thiam[a]

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Applied Radiation and Isotopes 210 (2024) 111364

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Measurements of the absolute gamma-ray emission intensities from the decay of ^{166}Ho

M. Capogni[a,*], A. Fazio[a], M. Vaccaro[b,1], P. De Felice[a]

[a] ENEA Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti (INMRI), Centro Ricerche Casaccia, I-00123, Rome, Italy
[b] University of Rome "La Sapienza", Departments of Physics, P.le A. Moro 2, I-00185, Rome, Italy

Applied Radiation and Isotopes 164 (2020) 109237

Contents lists available at ScienceDirect

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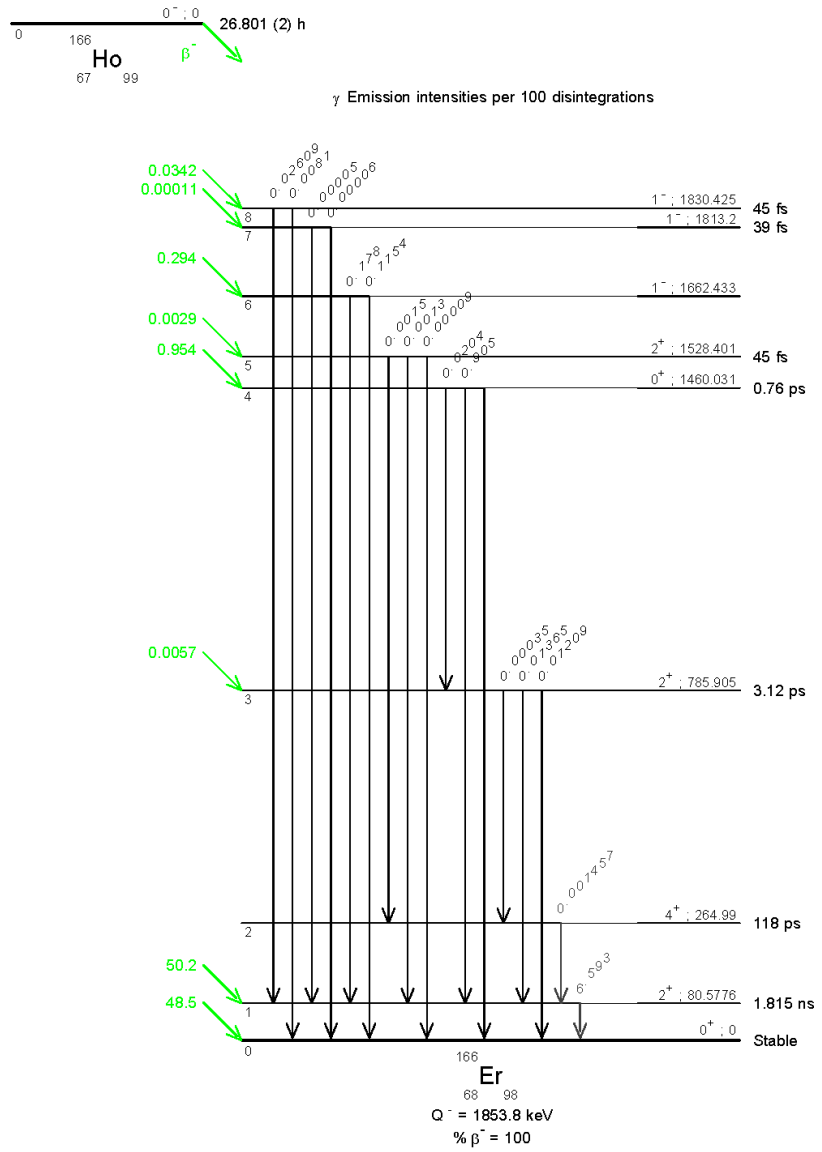
Primary standardization and determination of gamma ray emission intensities of Ho-166

I.M. Yamazaki, M.F. Koskinas *, D.S. Moreira, R. Semmler, F. Brancaccio, M.S. Dias

Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP, Av. Prof. Lineu Prestes 2242, 05508-000, São Paulo, SP, Brasil

Level scheme

- Gamma placements (17) based on level scheme proposed by Ardisson et al. (1992).
- Intensity balance: total sum of all transition energies Q_{Calc} = 1853.0(21) keV, mass-based Q-value Q_{Tot} = 1853.8(8) keV.
- Level energies spins, parities, half-lives, transition multipolarities, and mixing ratios taken from ENSDF Adopted Levels and Gammas.
- Level at 1813 keV not included by ENSDF, but proposed by Ardisson et al. and adopted in both previous and present DDEP evaluations.



Evaluation methodology

- SAISINUC (Dulieu, et al 2008) used to organize the evaluation results, most calculations done in Excel.
- Average values calculated with Limitation of Relative Statistical Weights Method (Browne 1998).
- Internal Conversion Coefficients: BrICC (Kibédi, et al. 2008) used with Frozen Orbital Approximation.
- X-ray and Auger electron emission intensities: 2013 version of the EMISSION code (Schönfeld and Janssen 2000).
- Q_{Tot} calculated from mass evaluation data of Wang, et al. (2021).
- Fluorescence yields calculated from data of Schönfeld and Janssen (1996).
- X-ray and Auger electron energies: Schönfeld and Rodloff (1999) and Schönfeld and Rodloff (1998), respectively.
- log *ft* values and mean β^- energies: BETASHAPE code (Mougeot 2017, 2023).
- All uncertainties quoted as corresponding to a single (k=1) uncertainty interval.

Half-life

- Values with < 2 decimal places not considered because of availability of higher quality data
- Daniel and Kaschl rejected by LWEIGHT by the Chauvenet principle
- Abouzi, et al. value removed from data set due to very short measurement time; Ramaniah et al. not included because uncertainty was clearly underestimated for technique used
- Fitzgerald and Unterweger (2020) taken as final value from NIST
- Multiple values in Bobin, et al. from same laboratory were combined to give a single value for each
- Recommended half-life is the weighted mean of six values remaining from original data set after removal of outliers, etc.
- The uncertainty from LWEIGHT was increased to the minimum uncertainty in the data set (LNHB value).

Reference	Year	$T_{1/2}$ (h)
Bothe	1946	27.3(5)
Grant and Hill	1949	26.8(4)
Antoneva et al.	1950	27.5(5)
Cork et al.	1958	26.9(1)
Hoffman	1963	27.0(2)
Daniel and Kaschl	1966	26.74(5)
Nethaway and Missimer	1968	26.79(3)
Ryves and Zieba	1974	26.83(3)
Ramaniah et al.	1976	“27 ± 0.04”
Abouzi et al.	1989	26.827(5)
Bobin, et al. (CMI)	2019	26.828(28)
Bobin, et al. (LNHB)	2019	26.811(20)
Bobin, et al. (NPL)	2019	26.797(26)
Unterweger and Fitzgerald	2020	26.794(23)
Recommended		**26.808(20)**

Evaluation of P_γ

- Five primary sources: Ardisson, et al. (1992); Coursey, et al. (1994); Bobin, et al (2019) – 3 sets; Yamazaki, et al. (2020); and Capogni, et al. (2024).
- Transition energies taken from Ardisson, et al.
  - Same source as previous DDEP evaluation
  - Other values available (e.g., from ^{166}Tm EC decay) but not better
- Ardisson, et al. is the only set not reporting absolute measurements directly from their own measurements
  - Normalization based on 1962 β-γ measurements that were done with NaI(Tl) detectors by Cline, et al.

Evaluation of P_γ : Step 1

- Evaluation of normalization factor
  - Calculate absolute probability of 1379.4 keV gamma without Ardisson value.
  - Ardisson data adjusted by removing Cline normalization and applying new normalization factor
  - Ardisson data did not include uncertainty on Cline data, uncertainty was assumed to be associated only with relative probabilities (about 1 %)
  - Uncertainty on relative probabilities combined with uncertainty on new normalization factor

Reference	$P_{\gamma,\ 1379.4\ keV}$
Ardisson, et al. (1992)	-
Coursey, et al. (1994)	-
Bobin, et al. (2019) (CMI)	0.904(20)
Bobin, et al. (2019) (LNHB)	0.896(13)
Bobin, et al. (2019) (NPL)	0.9051(69)
Yamazaki, et al. (2020)	0.904(11)
Capogni, et al. (2024)	0.902(29)
Recommended	**0.905(6)**

Evaluation of P_γ : Step 2

- LWEIGHT used to absolute probabilities for each gamma ray in data set, including renormalized Ardisson, to arrive at recommended values
- Examples – Excel spreadsheet calcs
  - LWEIGHT calculation sheets for gamma rays
  - Collection of recommended values for total intensity calculations
- Alternative approach (current ^{212}Pb evaluation)
  - Fewer absolute measurements, normalizations done on different members of decay chain
  - Evaluate normalization factor from small data set
  - Calculate relative intensities for all data sets
  - Determine LWEIGHT-calculated relative intensities
  - Apply evaluated normalization factor to average relative intensities

Evaluation of P_γ : Notes

- Data for 184.5 keV gamma ray are discrepant
  - Only two values: Ardisson and Bobin (NPL)
  - Uncertainty on Ardisson value was about 10 %, while Bobin (NPL) uncertainty was 4.8 %.
  - Bobin (NPL) value chosen as recommended value on the basis of being a better number
- Data for 674.2 keV gamma were also discrepant
  - LWEIGHT weighted average taken as recommended value
  - Uncertainty expanded to include the most precise value (Ardisson)
- Data are lacking for gammas above 1250 keV – more measurements needed

Special note: 1460 keV γ-ray (E0 transition)

- Unique transitions between two 0^+ states
- Photons have spin = 1, so γ-ray emission with $\Delta l = 0$ is forbidden
- Transition is possible through interactions between nucleus and atomic electrons
  - Internal conversion
  - Pair production
- Transitions are characterized by 100 % electron emission, no photons

Special note: 1460 keV γ-ray (E0 transition), pt 2

- Transition intensity calculated from experimental electron data
- Grigoriev, et al. (1974): $I_{K,1460} / I_{K,1379} = 0.2(1)$
- Evaluated $I_{\gamma,1379} = 0.905(6)/100$ decays → $1.356(21) \times 10^{-3}\ e_K / 100$ decays
- $I_{K,1460} = 2.7(14) \times 10^{-4}\ e_K/100$ decays
- $\alpha_K > \alpha_{l,m,\ldots}$, so considering uncertainty, $\alpha_k \approx \alpha_T$

Internal conversion, etc.

- Multipolarities taken from ENSDF Adopted Gammas Table
- Experimental mixing ratios used when possible (e.g., from γ–γ angular correlation measurements), otherwise equal mixing between multipolarities (e.g., M1+E2) is assumed (δ = 1)
- ICCs calculated with BrICC (Frozen Orbital Approximation)
- Atomic data (fluorescence yields, x-ray and Auger intensities) calculated with EMISSION in SAISINUC
- Consistent with previous evaluations; x-ray data consistent also consistent with new measurements (Bobin (all labs)).

β^- branching

- (New) recommended gamma-ray emission probabilities and ICCs used to calculate total intensity for each transition: $I_{tot} = P_\gamma (1+\alpha_{Tot})$
- Calculate sum of total intensity feeding into and de-exciting each level
- "Extra" intensity that de-excites must be due to additional intensity fed directly into level by beta decay
- log *ft* and β_{avg} calculated with BetaShape within SAISINUC
- Example: Excel spreadsheet calculations

Final Steps

- Copy data to SAISINUC, validate data entries, run consistency checks
- Input references
- Write Comments file (I do this concurrently with evaluation to track what I actually do, along with keeping separate notebook)

Conclusions

- Demonstrated process used for recent Ho-166 evaluation
- Revealed need for additional gamma-ray measurements at higher energy (> 1 MeV)
- Personal notes
  - Data validation in copying values into spreadsheets and into SAISINUC is critical and is perhaps the area that I need to improve on the most
  - Keep a notebook that details what publications were considered (perhaps short summary?), steps taken, decisions made, etc. since an evaluation is rarely performed within one block of time
  - Some knowledge of nuclear structure is very helpful in making some decisions or judging consistency