A Simple Tool for Photonuclear Activation Calculation

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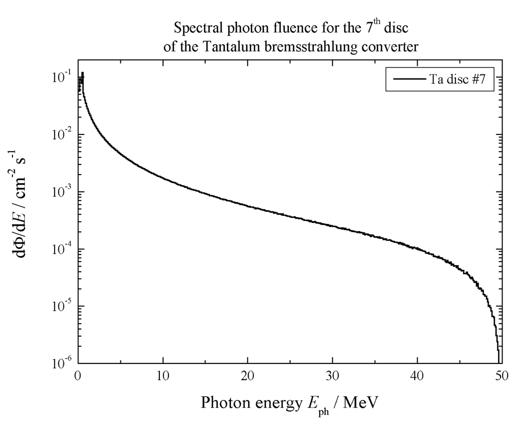
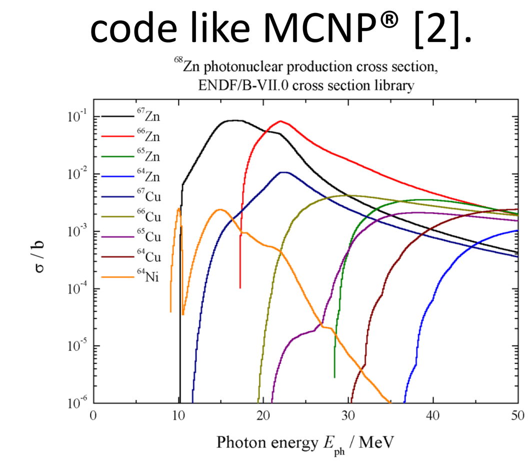
Motivation

- The beta emitters ^{47}Sc and ^{67}Cu are promising radionuclides for cancer therapy due to decay properties similar to ^{177}Lu and convenient half lifes.
- Besides charged-particle reactions, a large scale production is possible by photonuclear reactions like ^{48}Ti(γ,p)^{47}Sc and ^{68}Zn(γ,p)^{67}Cu.
- Bremsstrahlung radiation of a high-current electron accelerator can produce the radionuclides of interest but can also cause the production of unwanted radionuclides in the target material, Bremsstrahlung converter and structural materials.
- To the knowledge of the authors, no simple tool (like IAEA Medical Isotope Browser [1] for charged-particle reactions) exists for photonuclear reactions.

Procedure

- 1st step: Calculation of spectral photon fluence with a radiation transport code like MCNP® [2].





- 2nd step: Normalization to beam current, calculation of reaction rates.
- 3rd step: Solution of Bateman equations [3] with additional terms for photonuclear reactions: iterative calculation of atom densities in small time steps, calculation of activity concentrations and specific activites.

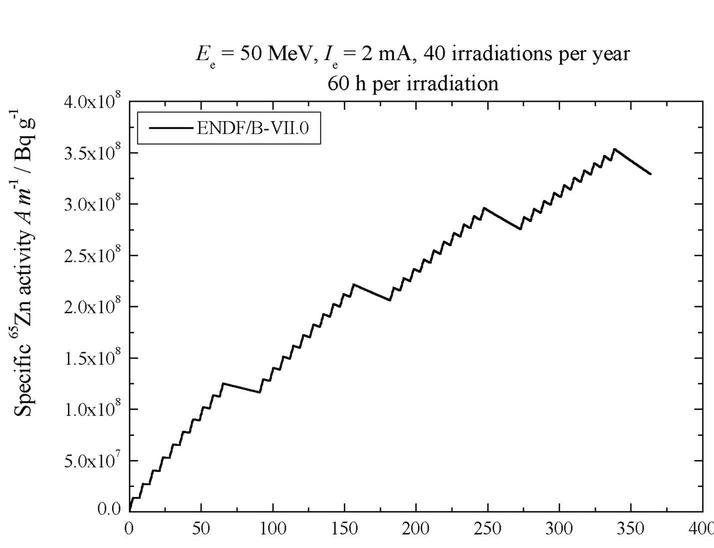
Tool for Activation Calculation

- Originally intended for the analysis of activation experiments and yield measurements.
- Derived from a tool for neutron activation calculation that proved to be useful in a decommissioning project [4].
- Limited number of materials (high Z converter materials, Mo, Zn and Ti as target materials, structural materials are currently included).
- Planned irradiation facility foresees a 50 MeV electron LINAC (More information on Poster #23 “Design and Feasibility Study for the Large-Scale Production of ^{67}Cu Using a High-Power LINAC Electron Beam”). Implemented reactions are therefore limited to ≤ 50 MeV photon energy.
- Implemented as VBA (“big Excel macro”), compatible with LibreOffice Basic

Fields of Applications

Co-production of ^{65}Zn

- Irradiation of a ^{68}Zn sample will cause the production of gamma-emitter ^{65}Zn with $T_{1/2}$ = 244 d.
- High yield of the ^{68}Zn(γ,3n)^{65}Zn reaction in spite of the high reaction threshold.
- Repeated use of enriched material leads to accumulation of activity.



- While analytical solution of Bateman equations is possible, the numerical calculation is convenient for repeated cycles of irradiation and cool-down.

Evolution of Zn Isotopic Ratio

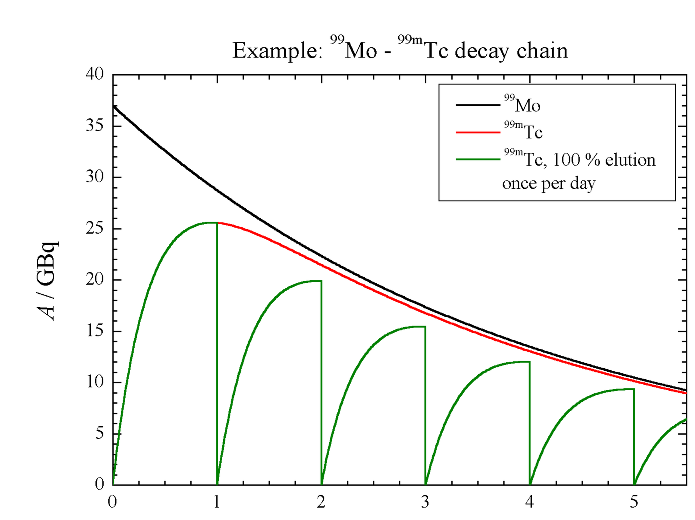
- Both reactions for the production of radioactive and stable isotopes are included.
- Code allows the calculation of changes of the isotopic ratio of the irradiated material. Here: assumption of a 360 d irradiation of isotopically pure ^{68}Zn.

Isotope	Atom density n / atoms cm^{-3}
Start of irradiation	
^{68}Zn	6.575 × 10^{22}
End of irradiation	
^{68}Zn	6.568 × 10^{22}
^{67}Zn	4.59 × 10^{19}
^{66}Zn	2.05 × 10^{19}
^{65}Zn	2.82 × 10^{17}
^{64}Zn	5.28 × 10^{16}
^{63}Zn	4.87 × 10^{9}
^{62}Zn	1.48 × 10^{9}

- Depletion of expensive enriched material plays only minor role.

^{99}Mo – ^{99m}Tc Decay Chain

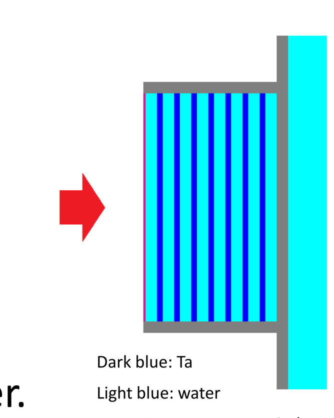
- Activation tool includes relevant radionuclides with $T_{1/2} \geq 1$ min.
- Care was taken to include radioactive decay data and branching ratios for decay chains.



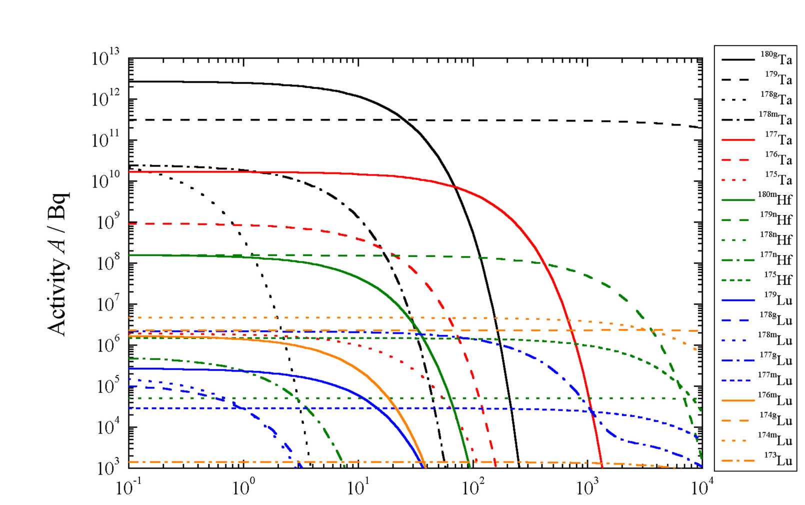
- Code replicates well the “teaching book” example of the decay chain of a 37 GBq (1 Ci) ^{99}Mo-^{99m}Tc generator.

^{nat}Ta Activation

- Concept of a water-cooled Tantalum Bremsstrahlung converter divided into seven discs with thickness 0.5 mm each to improve heat transfer.



- Activity estimation for 7th disc (highest photon fluence) after 360 d of irradiation is shown.



- Activity is dominated by ^{180g}Ta ($T_{1/2}$ = 8 h) and ^{179}Ta ($T_{1/2}$ = 1.82 y). Small activity of radionuclide with longest half-life (^{178n}Hf, $T_{1/2}$ = 31 y).

Reaction Cross Section Data

- Calculated activities strongly depend on quality of cross section data libraries. Reaction cross section data can strongly deviate in different libraries.
- ENDF/B-VII.0: Only available for selected materials. No reaction data for production of isomeric states. Some data sets limited to 30 MeV. Evaluation process includes experimental data.
- TENDL: Very comprehensive data set. Indications (i.e. [5]) of underestimation of charged particle emission by medium and heavy nuclei. Severe underestimation of ^{68}Zn(γ,p)^{67}Cu yield!
- Mix-and-match: Use of ENDF/B-VII.0 if available, JENDL/PD-2016.1 for light nuclei, TENDL for majority of reactions.

Conclusions and Outlook

- A code for photonuclear activation calculation is currently developed at HZDR.
- Early activity estimation of unwanted byproducts can support the design of a planned facility for the photonuclear production of medical radioisotopes.
- Work-in-progress: increase of the number of available materials with focus on structural materials (steel components, major concrete components).
- Coupling with a neutron activation code to account for photoneutrons is discussed.

Acknowledgements

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References

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