Applications and reactions on unstable nuclei

WANDA 2022

Jo Ressler with:

P. Bedrossian, R. Hoffman, V. Mozin, W.-J. Ong, A. Ratkiewicz, M. Robel
Reactions on unstable nuclei require pretty intense conditions for applications...

- Unstable nuclei considered here have decay half-lives < 1e6 years

- With shorter half-lives, application must produce unstable isotope through one reaction, and destroy it in another
  - Second-order reaction
  - Must produce a significant quantity of intermediate radioactive isotope
  - Production/destruction in same locale

- One of two conditions:
  - High flux of particles or photons + residence time or
  - Very high flux of particles or photons (extreme conditions)

- Considered nuclear data needs
  - Most applications require radiochemistry, material science, engineering...
  - Data can be measured or theoretical, as long as it is accurate – applications care about the data (preferably in an evaluated library)
Medical physics utilizes reactions on unstable isotopes for production of (some) therapeutic nuclides

- **Diagnostic and therapeutic applications**
  - Diagnostic: imaging; PET, SPECT *example:* $^{68}$Ga
  - Therapeutic: targeted energy deposition *example:*$^{177}$Lu
  - Theragnostic: Two isotopes of the same element for diagnostic and therapeutic *examples:* $^{64}$Cu-$^{67}$Cu, $^{43,44}$Sc-$^{47}$Sc

- **Wide variety of isotopes produced**
  - Proton-rich in cyclotron, synchrotron, accelerator: $^{18}$F, $^{67}$Ga, $^{123}$I, $^{201}$Tl
  - Neutron-rich in a fission reactor: $^{99m}$Tc, $^{131}$I, $^{166}$Ho, $^{177}$Lu

- **High-flux reactor-produced isotopes on intermediate products**
  - $^{186}$W(n,γ)$^{187}$W(n,γ)$^{188}$W $\rightarrow$ $^{188}$Re
  - $^{192}$Os(194Ir), $^{164}$Dy(166Ho)
  - $^{175}$Lu(n,γ)$^{176}$Lu(n,γ)$^{177}$Lu

- **Data are lacking for neutron reactions on unstable nuclei**
  - Need to understand production options and purity issues

---

Our current generation of nuclear power reactors has known reactions, near future has unknowns

- Reactions on radioactive isotopes occur with Light Water Reactors (LWRs)
  - Actinides, neutron capture and fission, Am and Cm in spent fuel
  - Fission product generation, neutron capture example $^{135}$Xe, $^{149}$Sm poisons

- Higher burnup under consideration
  - Currently ~45 GWd/MTU, increasing to 75 – 80 GWd/MTU
  - Longer irradiation periods shift neutron spectrum, reaction rates
  - Capture effects on fission products and structural materials will increase

- Accident Tolerant Fuels: changes to fuel including enrichment, cladding materials, pellet design...

- Analytical models may not be fully validated in new operating regimes, lack data
  - Affects predictive capability
  - Licensing and control, based on material loading/unloading, reactor operation

*Burnup credit isotope $^{148}$Nd depends on fission and capture reactions*
Reactor monitoring applications will be challenged with unknowns

- Safeguards, forensics, and non-proliferation activities have less access to reactor details

- Interdicted or environmentally sampled materials
  - Unknown operating parameters from known reactor
  - Unknown reactor (and unknown operating parameters)

- Research with $^{133}\text{Cs}/^{135}\text{Cs}$ ratios in spent fuel
  - Predicted to vary linearly with neutron flux over typical LWR power range
  - Validation tests with BR3 reactor

---

**Diagram:**

- $^{133}\text{Cs}$: 100%
- $^{134}\text{Cs}$: 2\text{y}
- $^{135}\text{Cs}$: 2e6\text{y}
- $^{133}\text{Xe}$: 2d/5d
- $^{134}\text{Xe}$: 10%
- $^{135}\text{Xe}$: 9h
- $^{133}\text{I}$: 21h
- $^{134}\text{I}$: 53m
- $^{135}\text{I}$: 7h

**Graph:** Modeled and Experimental

- Data ($\mu \pm 2\sigma$)
- Model
- Model $\pm 10\%$

Systematic error due to nuclear data or other effects?
Next generation energy reactors will utilize new materials and processes

- Small Modular Reactors (<300 MWe)
  - iPWR, MSR, HTGR, LMFR... 50+ concepts
  - Russia, China, US, Canada, South Korea, France

- Alternate fuels
  - High Assay Low Enriched Uranium (HALEU)
  - Different compounds and material states (e.g. liquid or sintered)
  - Burnable poisons

- New operating regimes
  - Higher burnup
  - Dynamic fuel movement through the core (e.g. liquids or pebble-bed)

- Closed, semi-closed fuel cycles; Mixed Oxide (MOX), Fast breeder reactors

- Will involve different isotopes, environments, neutron energies
  - Analytical models may be unvalidated or incomplete
  - Safeguards challenges

New York Times, 11/5/2021
Stockpile Stewardship Program utilizes a large number of reactions on unstable nuclei

- Understanding our test history
  - Fission product yields, reactions on fission products
  - Radiochemical tracers

- Knowledge of production and destruction networks needed
  - Competing capture and (n,2n) reactions

\[ (n,\gamma) \quad (n,2n) \quad (n,n') \]

- \( ^{94}\text{Zr} \) stable
- \( ^{95}\text{Zr} \)
- \( ^{96}\text{Zr} \) stable
- \( ^{94}\text{Y} \) 19 m
- \( ^{95}\text{Y} \) 10 m
- \( ^{96}\text{Y} \) 5.3 s
- \( ^{94}\text{Sr} \) 75 s
- \( ^{95}\text{Sr} \) 25 s
- \( ^{96}\text{Sr} \) 1.1 s
- \( ^{94}\text{Rb} \) 2.7 s
- \( ^{95}\text{Rb} \) 0.4 s
- \( ^{96}\text{Rb} \) 0.2 s

- \( ^{87}\text{Y} \) 87Y
- \( ^{88}\text{Y} \) 88Y
- \( ^{89}\text{Y} \) stable

- \( ^{137}\text{Y} \)
- \( ^{90}\text{Y} \) 909 keV
- \( ^{392}\text{keV} \)
- \( ^{675}\text{keV} \)

- \( ^{94}\text{Zr} \)
- \( ^{95}\text{Zr} \) stable
- \( ^{96}\text{Zr} \) stable
- \( ^{94}\text{Sr} \) 64 d
- \( ^{94}\text{Sr} \) 95Y
- \( ^{95}\text{Sr} \) 64 d
- \( ^{96}\text{Sr} \) 95Y
- \( ^{96}\text{Sr} \) stable

- \( ^{94}\text{Rb} \)
- \( ^{95}\text{Rb} \) 96Y
- \( ^{96}\text{Rb} \) 96Y

- \( ^{87}\text{Y} \)
- \( ^{87}\text{Y} \) (n,\gamma) \( ^{88}\text{Y} \)

- Cross Section (barns)
- Energy (MeV)

- Calculated
- Indirect meas.

- Calculated
- Indirect meas.

- Hoffman (HPSI)
- Escher (GSI)
Nuclear astrophysics provides non-terrestrial extreme environments

- Nucleosynthesis and origin of the elements
  - Astrophysical environments

- Trifecta:
  - Astronomical observations and laboratory measurements of stellar grains
  - Nuclear data
  - Astrophysics network calculations

- Energy produced through fusion, limited to elements less than Fe

- All heavier elements produced through later stages of stellar evolution or interstellar processes

- Extreme environments; reactions on unstable nuclei are common

https://www.science.org/doi/10.1126/science.aau9540
Isotope production processes

- **r-process, rapid neutron capture**
  - Large neutron flux, few seconds
  - Reactions very far from stability

- **s-process, slow neutron capture**
  - Low neutron flux, decades between neutron captures
  - Reactions along stability

- **i-process, intermediate between s- and r-**
  - Carbon-enhanced metal-poor stars
  - Neutron capture on unstable isotopes

- **P-nuclei, ~35 stable isotopes cannot be made in s-, r- process**
  - $\nu p$-process: neutrino winds in core collapse supernova create $p$-rich environment; proton capture and $(n,p)$
  - $\gamma$-process: core collapse supernova, photodisintegration of $s$-produced seed nuclei
Multiple applications utilize reactions on unstable nuclei

- High particle flux environment produces the unstable nuclei, which may undergo further (destruction) reactions

- Terrestrial applications with unstable isotopes are dominated by neutrons
  - Due to source availability; high flux of charged particles limited to astrophysical environments
  - Largely neutron capture, but other reactions such as \((n,2n)\) and \((n,p)\) can be important

- Lack of data, even close to stability
  - For thermal neutron reactor irradiation, Atlas of Neutron Resonances has limited (if any) data for one isotope off stability
  - Evaluated reaction data libraries, e.g. ENDF, often have no data

- Meeting data needs is a challenge: difficult measurements and theory