

# Applications and reactions on unstable nuclei

WANDA 2022

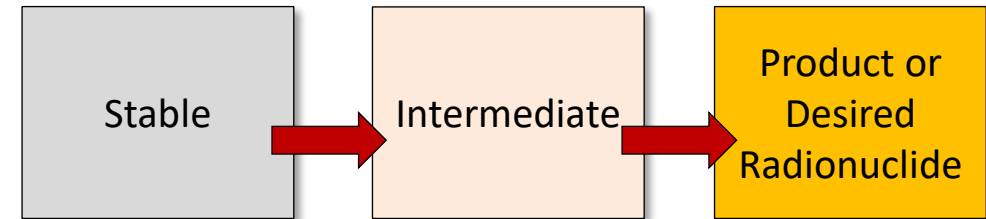
Jo Ressler with:

P. Bedrossian, R. Hoffman, V. Mozin, W.-J. Ong, A. Ratkiewicz, M. Robel



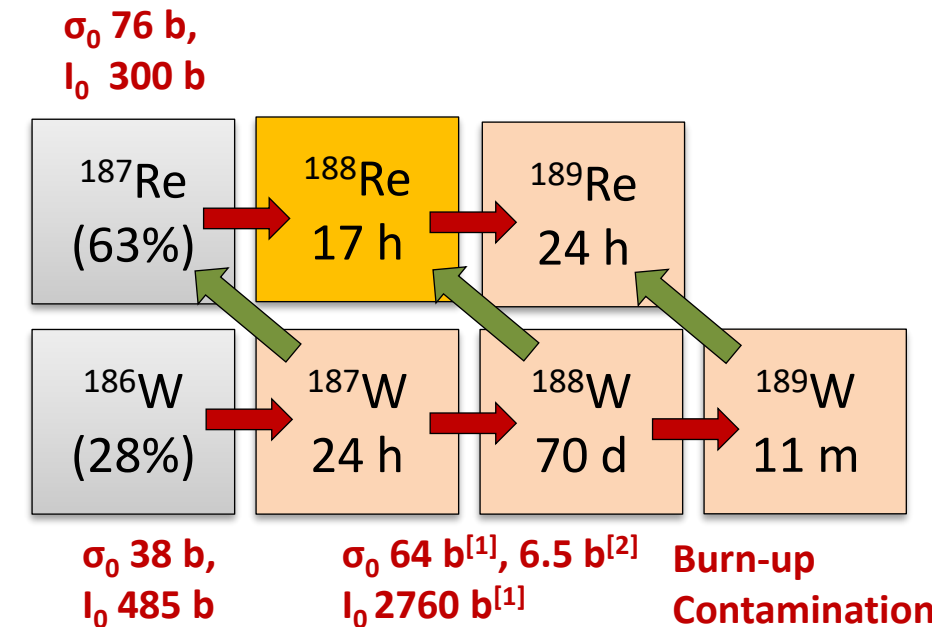
# Reactions on unstable nuclei require pretty intense conditions for applications...

- 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:  $^{68}\text{Ga}$*
  - Therapeutic: targeted energy deposition *example:  $^{177}\text{Lu}$*
  - Theranostic: Two isotopes of the same element for diagnostic and therapeutic *examples:  $^{64}\text{Cu}$ - $^{67}\text{Cu}$ ,  $^{43,44}\text{Sc}$ - $^{47}\text{Sc}$*
- Wide variety of isotopes produced
  - Proton-rich in cyclotron, synchrotron, accelerator:  $^{18}\text{F}$ ,  $^{67}\text{Ga}$ ,  $^{123}\text{I}$ ,  $^{201}\text{Tl}$
  - Neutron-rich in a fission reactor:  $^{99\text{m}}\text{Tc}$ ,  $^{131}\text{I}$ ,  $^{166}\text{Ho}$ ,  $^{177}\text{Lu}$
- High-flux reactor-produced isotopes on intermediate products
  - $^{186}\text{W}(n,\gamma)^{187}\text{W}(n,\gamma)^{188}\text{W} \rightarrow ^{188}\text{Re}$
  - $^{192}\text{Os}(^{194}\text{Ir})$ ,  $^{164}\text{Dy}(^{166}\text{Ho})$
  - $^{175}\text{Lu}(n,\gamma)^{176}\text{Lu}(n,\gamma)^{177}\text{Lu}$
- Data are lacking for neutron reactions on unstable nuclei
  - Need to understand production options and purity issues

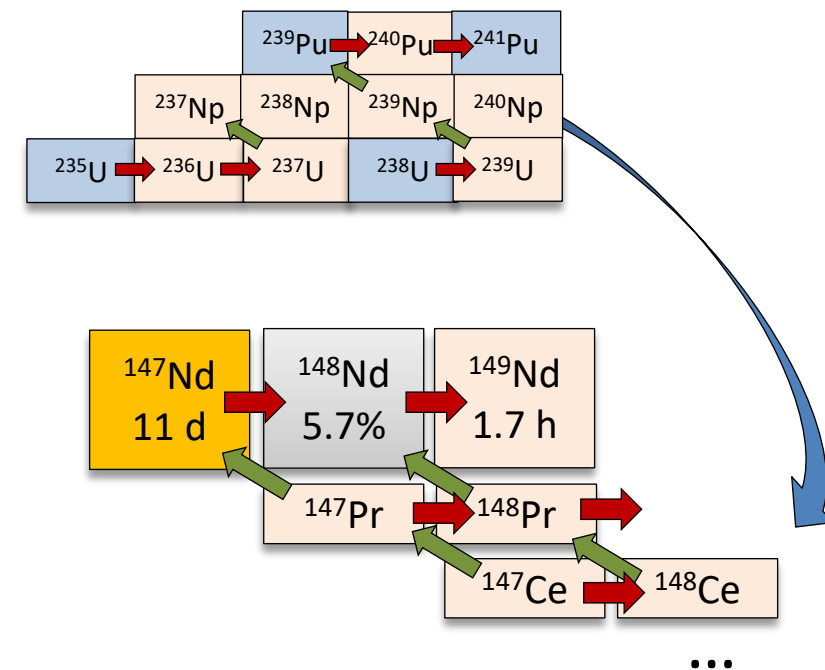


[1] 2018 Atlas of Neutron Resonances

[2] 2019 Appl. Rad. Iso, Esöz *et al*

# 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  $^{135}\text{Xe}$ ,  $^{149}\text{Sm}$  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

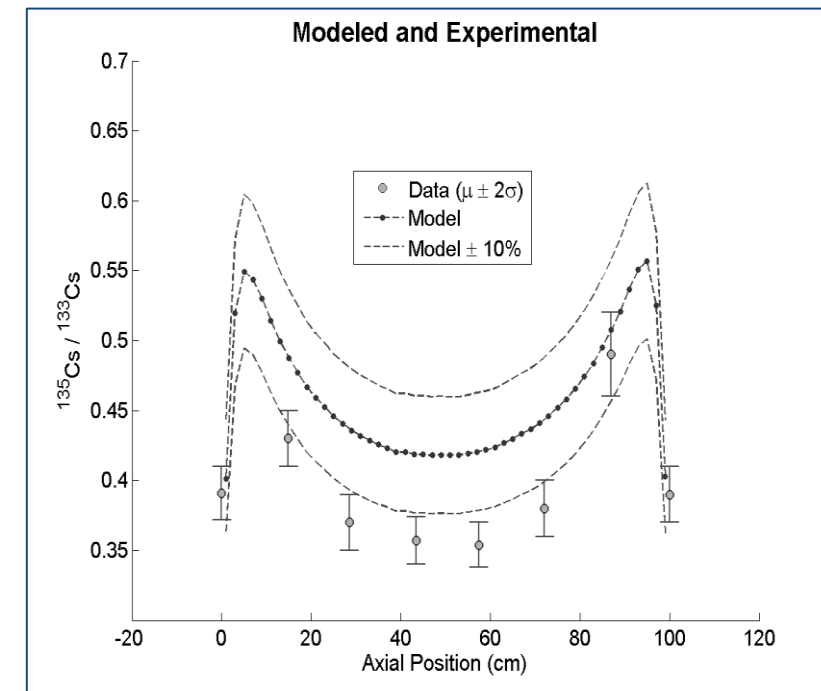
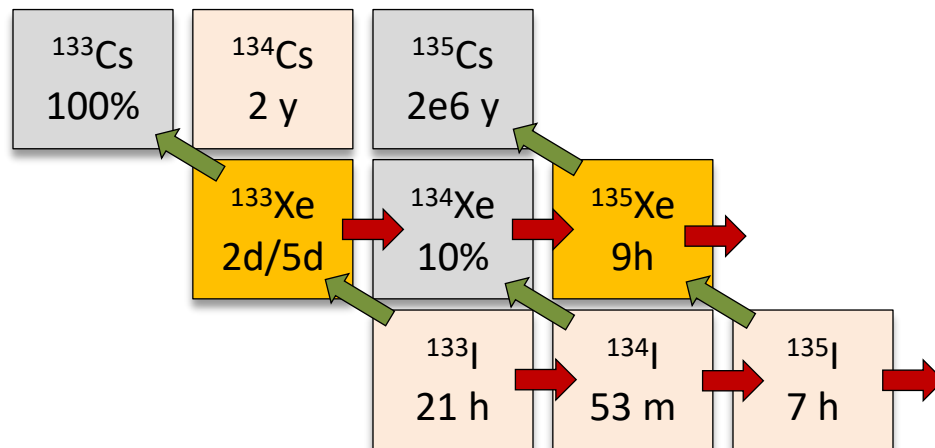


*Burnup credit isotope  $^{148}\text{Nd}$  depends on fission and capture reactions*



# 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



*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



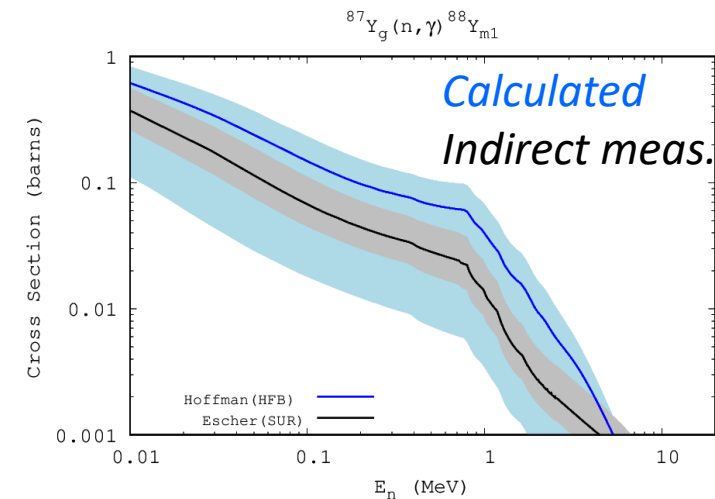
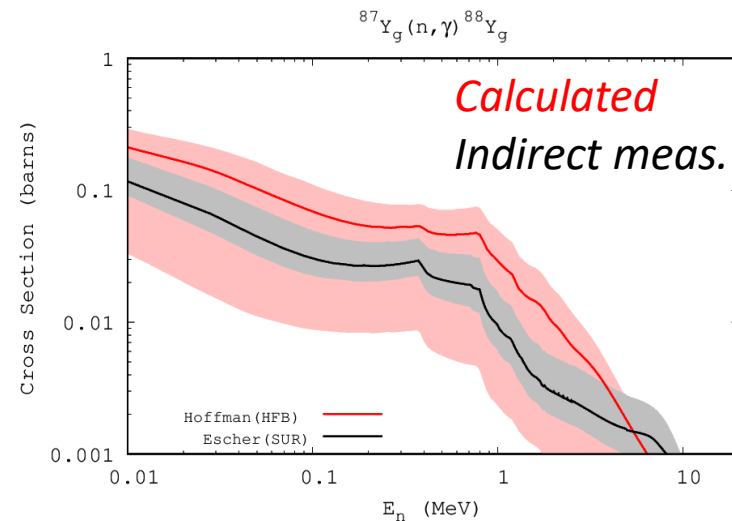
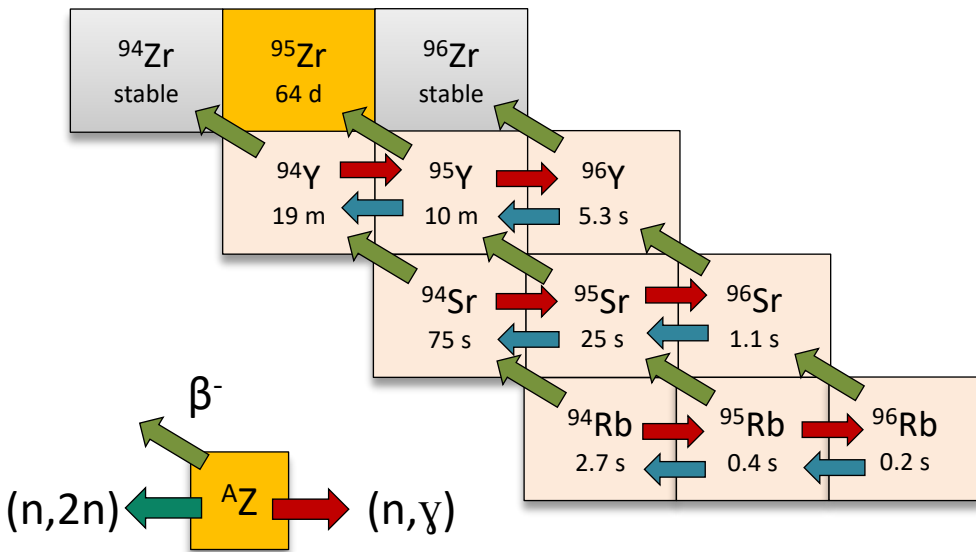
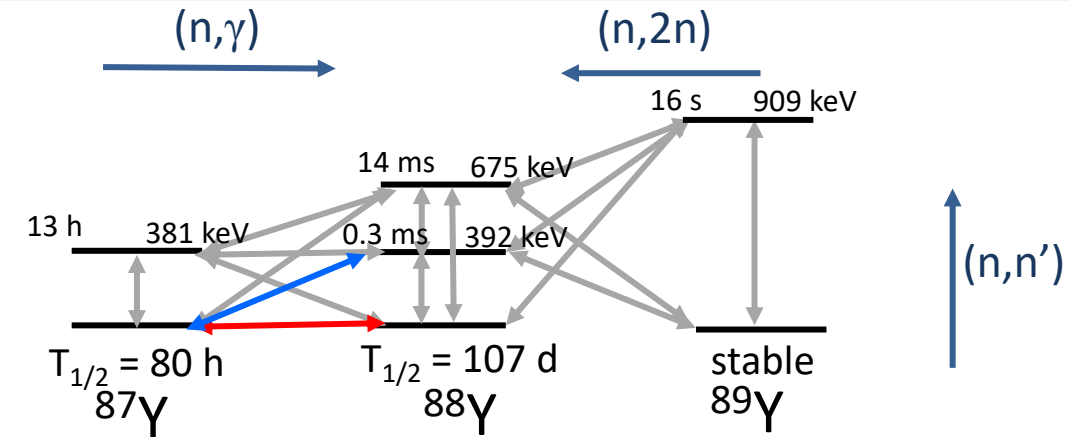
New York Times, 11/5/2021

<https://www.nytimes.com/2021/11/05/world/europe/russia-nuclear-power-climate-change.html>

*Akademik Lomonosov, Pevek, Russia*

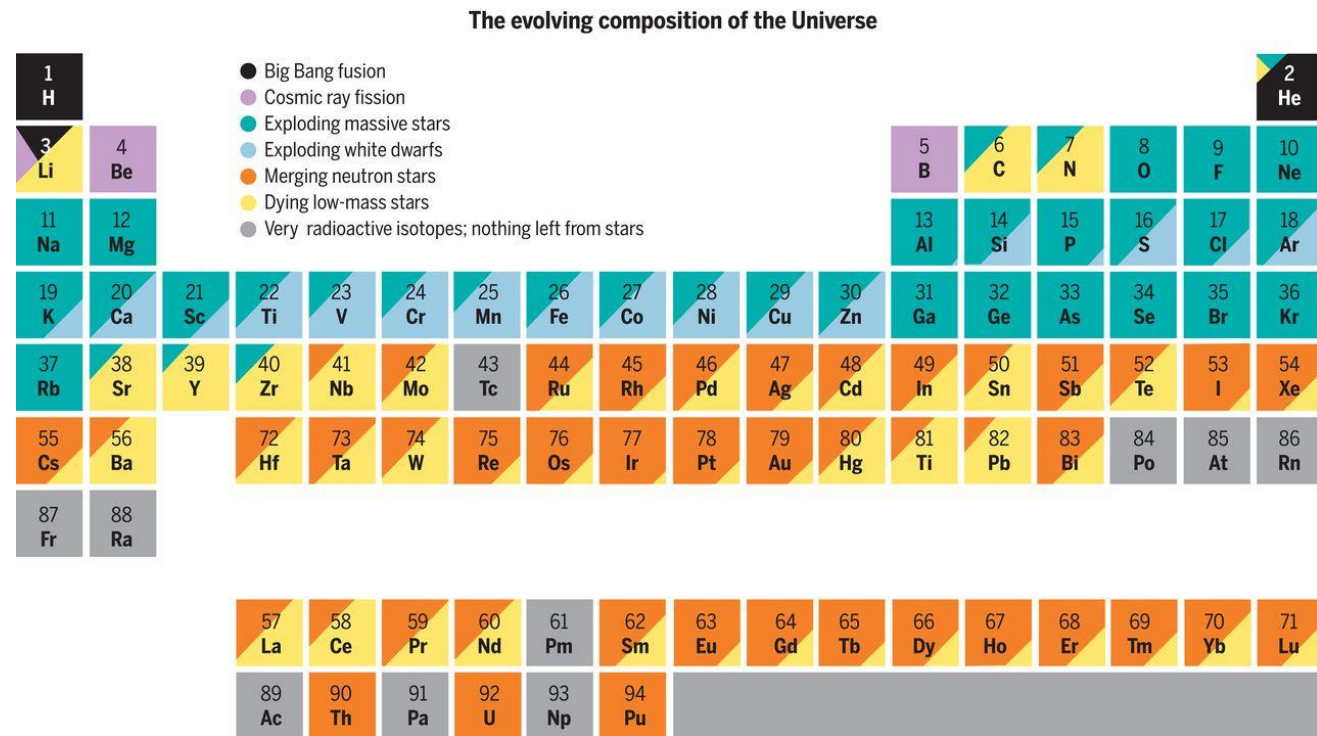
# Stockpile Stewardship Program utilizes a large number of reactions on unstable nuclei

- 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



# 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



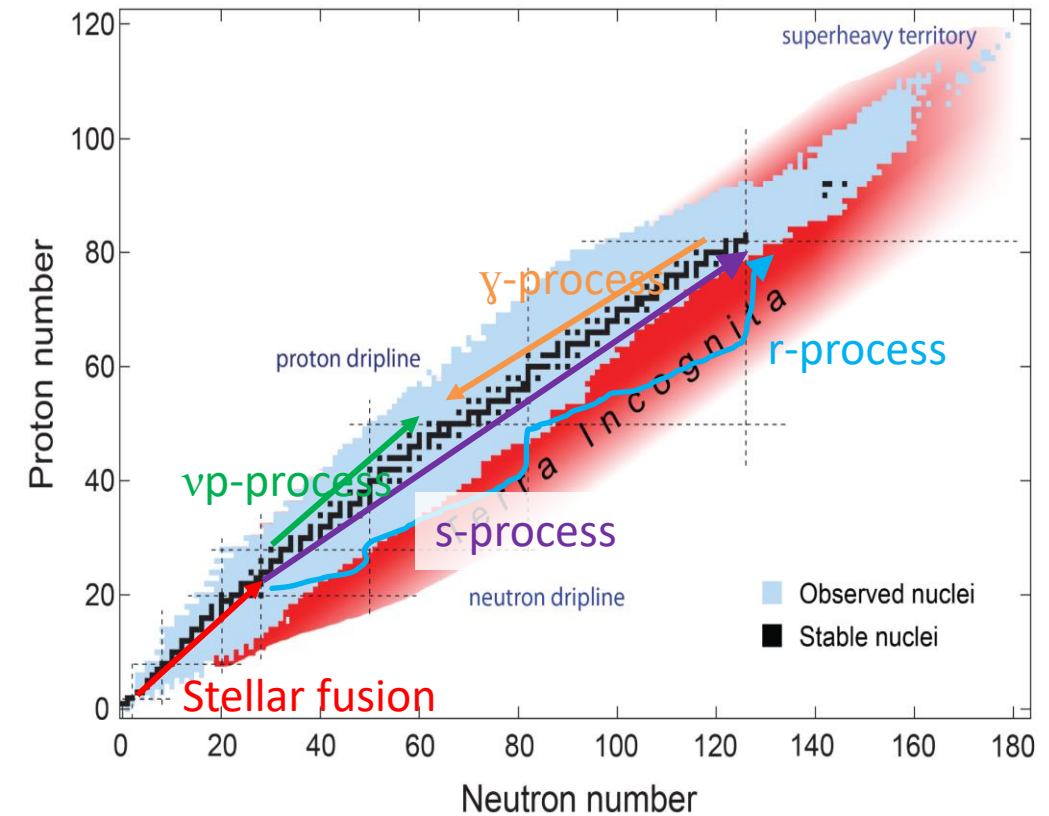
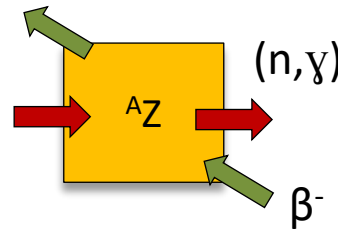
<https://www.science.org/doi/10.1126/science.aau9540>



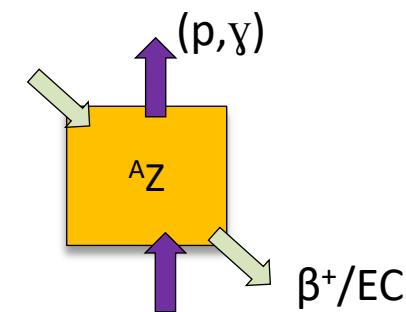
# Isotope production processes

- 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
  - vp-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

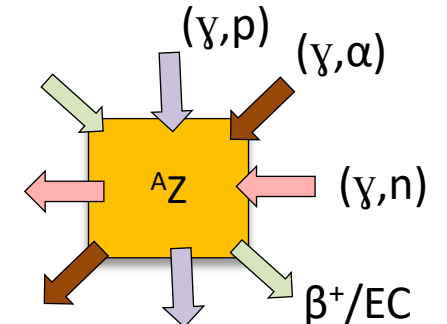
*r-, s-, i-process reactions:*



*vp-process:*



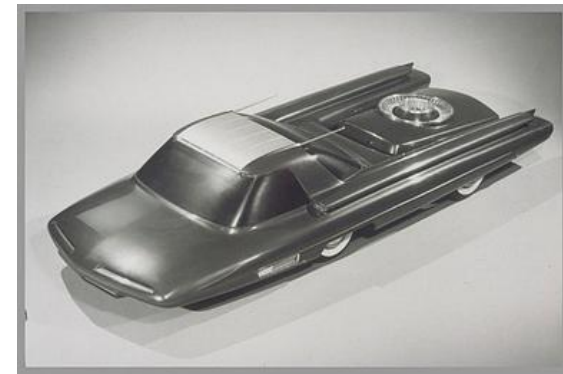
*γ-process:*



# 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

*1958 Ford Nucleon*



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<https://en.wikipedia.org/w/index.php?curid=685950>