Applications and reactions on unstable nuclei

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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


Stable → Intermediate → Product or Desired Radionuclide

- 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)

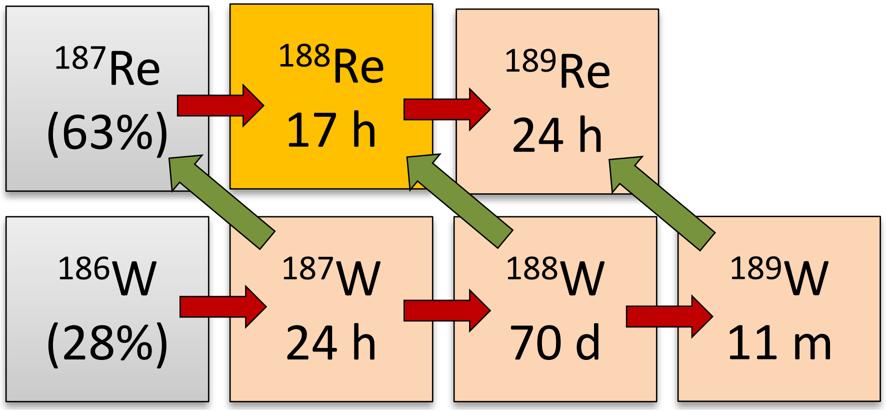
Medical physics utilizes reactions on unstable isotopes for production of (some) therapeutic nuclides

- 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
  - $^{186}\mathrm{W}(n,\gamma)^{187}\mathrm{W}(n,\gamma)^{188}\mathrm{W} \rightarrow {}^{188}\mathrm{Re}$
  - ^{192}Os(^{194}Ir), ^{164}Dy(^{166}Ho)
  - $^{175}\mathrm{Lu}(n,\gamma)^{176}\mathrm{Lu}(n,\gamma)^{177}\mathrm{Lu}$

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



σ_0 76 b,
I_0 300 b

^{187}Re (63%)

^{188}Re 17 h

^{189}Re 24 h

^{186}W (28%)

^{187}W 24 h

^{188}W 70 d

^{189}W 11 m

σ_0 38 b,
I_0 485 b

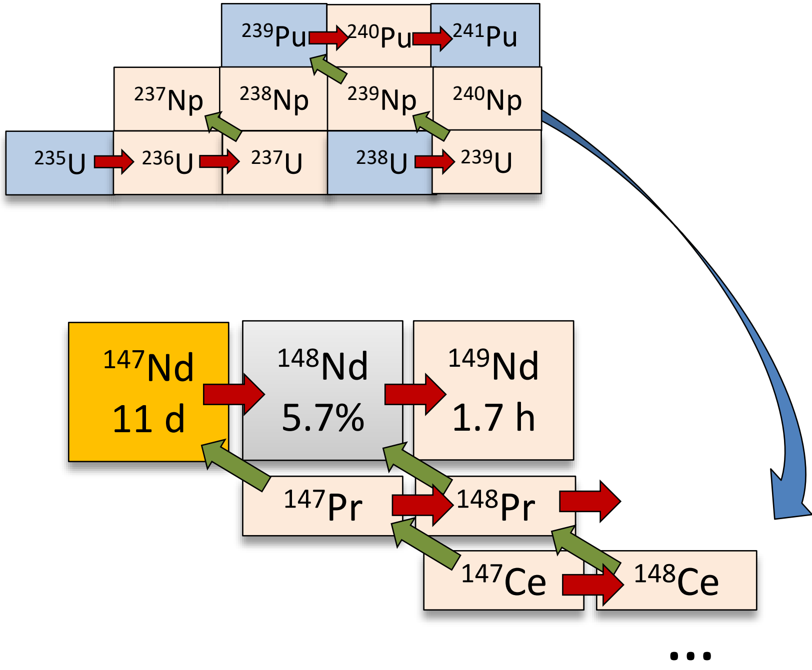
σ_0 64 b[1], 6.5 b[2]
I_0 2760 b[1]

Burn-up
Contamination

[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}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



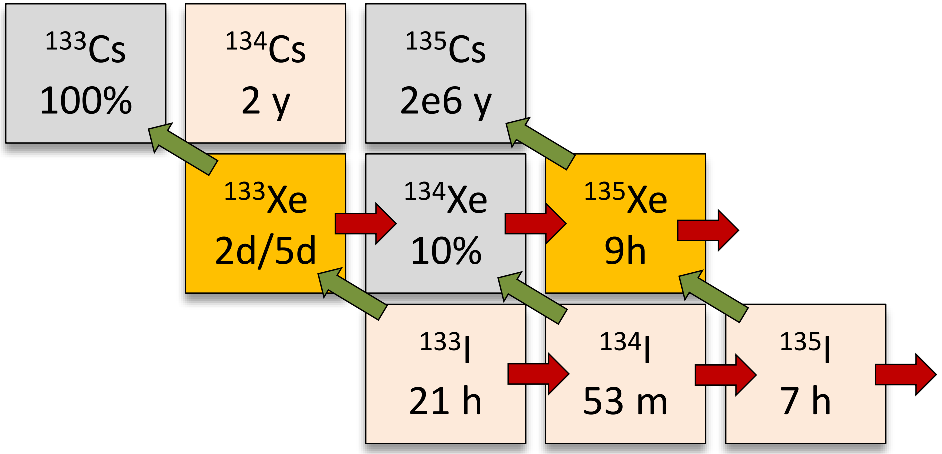
Burnup credit isotope ^{148}Nd depends on fission and capture reactions

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



^{133}Cs 100%

^{134}Cs 2 y

^{135}Cs 2e6 y

^{133}Xe 2d/5d

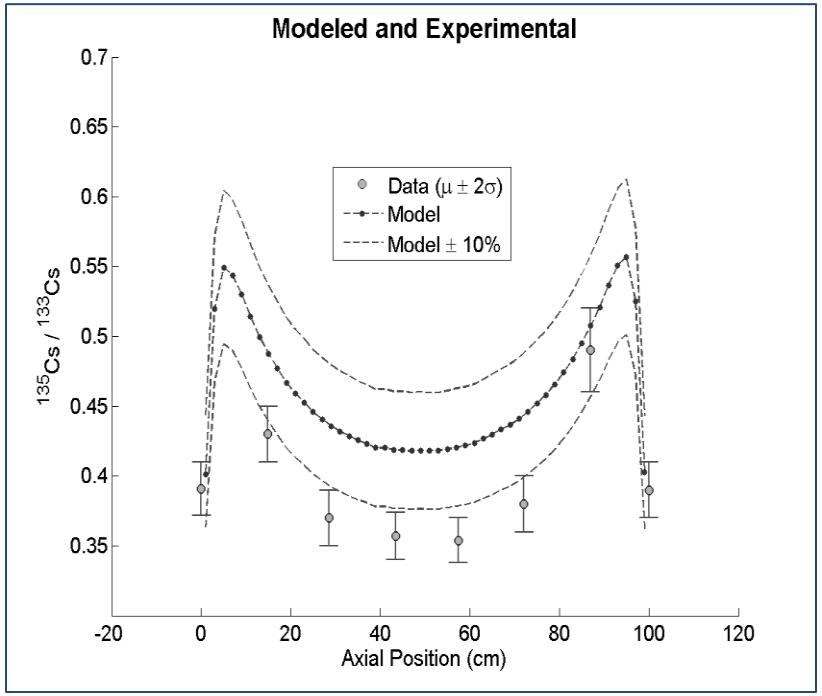
^{134}Xe 10%

^{135}Xe 9h

^{133}I 21 h

^{134}I 53 m

^{135}I 7 h



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

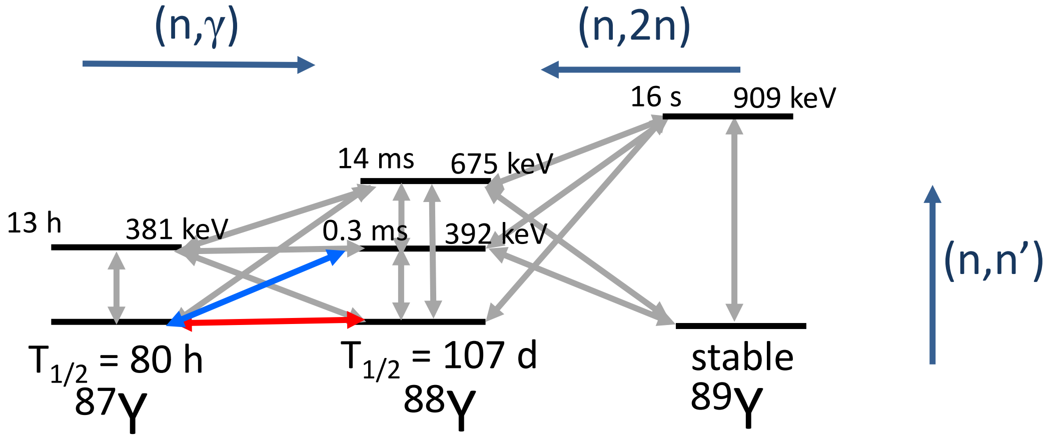
- Will involve different isotopes, environments, neutron energies
  - Analytical models may be unvalidated or incomplete
  - Safeguards challenges

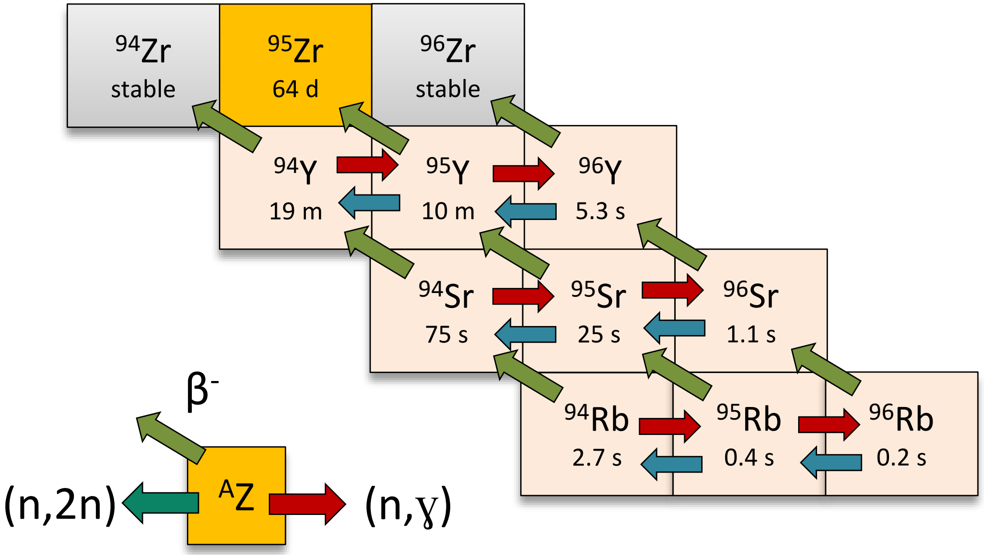
New York Times, 11/5/2021
https://www.nytimes.com/2021/11/05/world/europe/russia-nuclear-power-climate-change.html

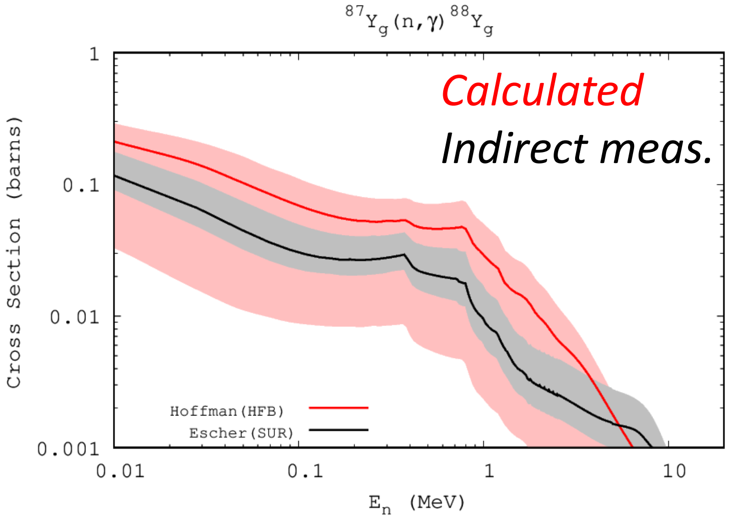
Akademik Lomonsov, Pevek, Russia

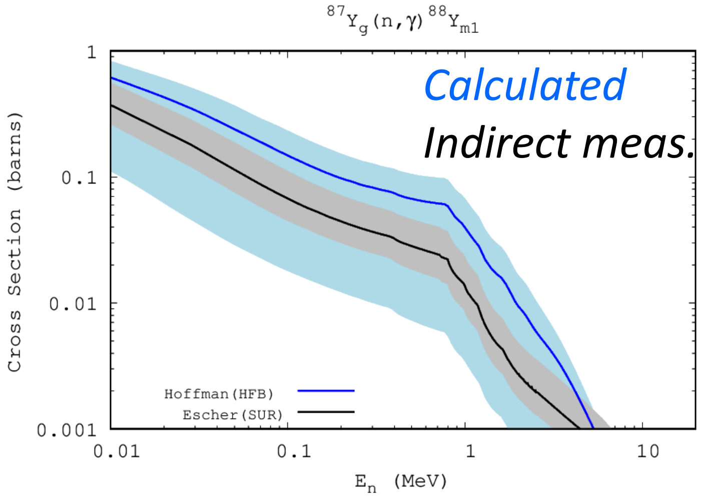
Stockpile Stewardship Program utilizes a large number of reactions on unstable nuclei

- 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






$^{87}Y_g(n,\gamma)^{88}Y_g$


$^{87}Y_g(n,\gamma)^{88}Y_{m1}$

Nuclear astrophysics provides non-terrestrial extreme environments

- Nucleosynthesis and origin of the elements
  - Astrophysical environments

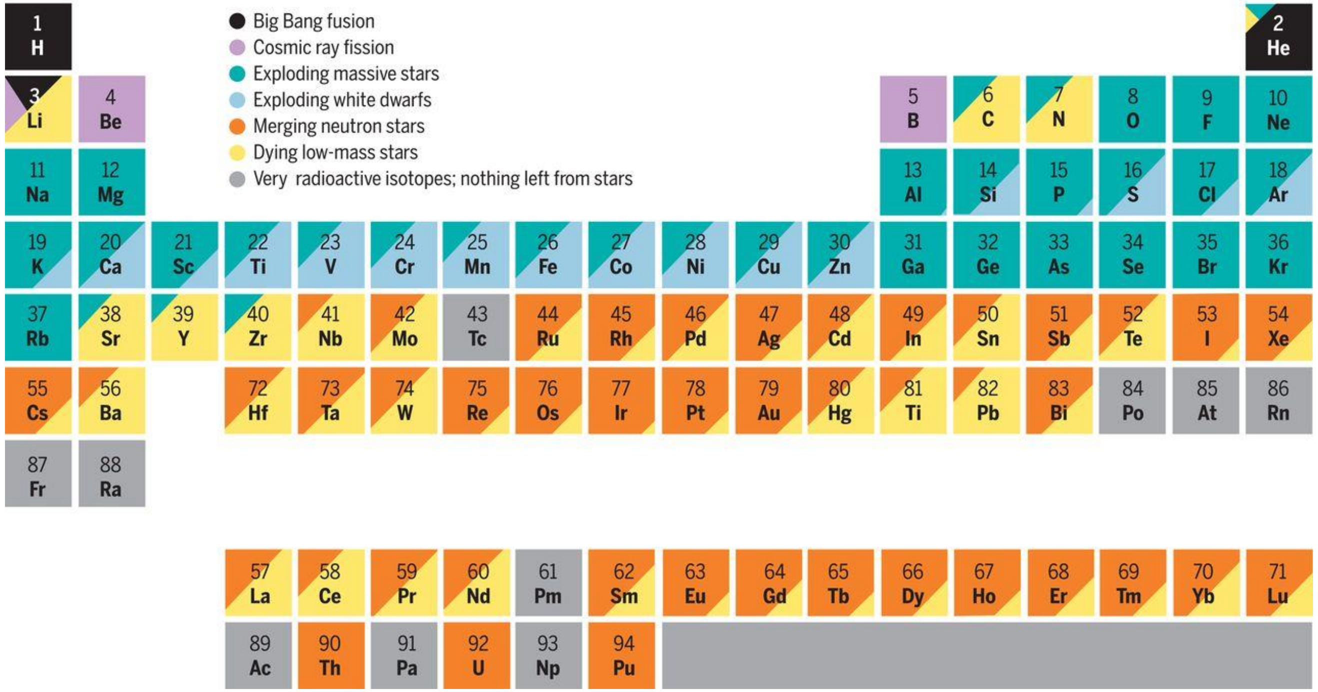
- Trifecta:
  - Astronomical observations and laboratory measurements of stellar grains
  - Nuclear data
  - Astrophysics network calculations

- 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

The evolving composition of the Universe

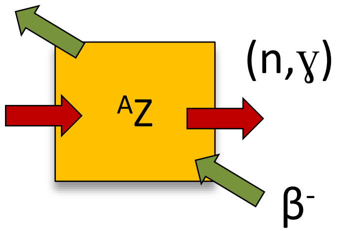


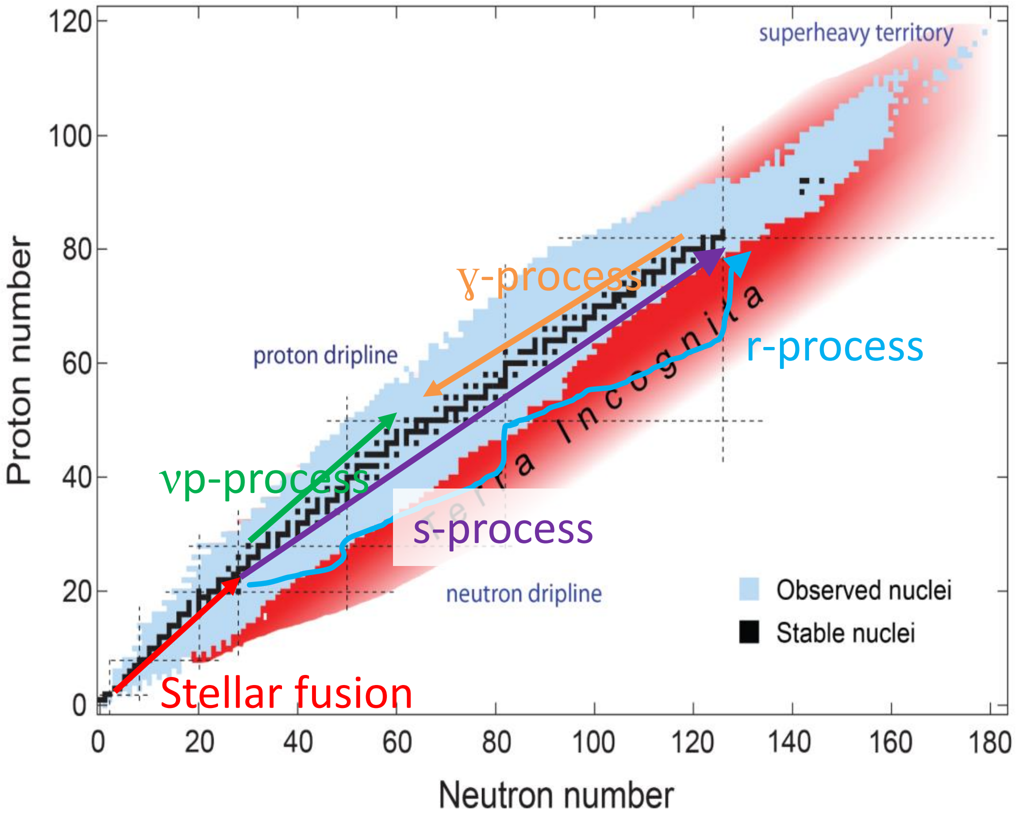
https://www.science.org/doi/10.1126/science.aau9540

Isotope production processes

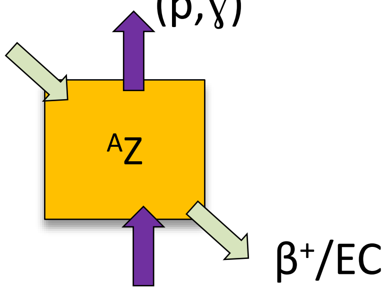
- r-process, rapid neutron capture
  - Large neutron flux, few seconds
  - Reactions very far from stability
- s-process, slow neutron capture
  - Low neutron flux, decades between neutron captures
  - Reactions along stability
- i-process, intermediate between s- and r-
  - Carbon-enhanced metal-poor stars
  - Neutron capture on unstable isotopes
- P-nuclei, ~35 stable isotopes cannot be made in s-, r- process
  - νp-process: neutrino winds in core collapse supernova create p-rich environment; proton capture and (n,p)
  - ɣ-process: core collapse supernova, photodisintegration of s-produced seed nuclei

r-, s-, i-process reactions:

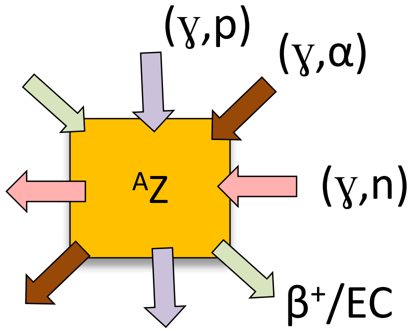




νp-process:



ɣ-process:



Multiple applications utilize reactions on unstable nuclei

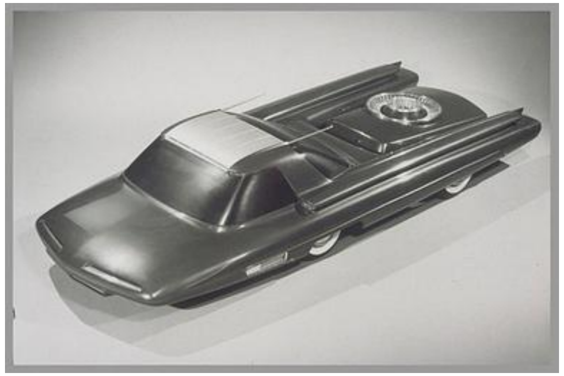
- High particle flux environment produces the unstable nuclei, which may undergo further (destruction) reactions

- 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