Brookhaven National Laboratory

U.S. DEPARTMENT OF ENERGY

TREND update: a TRi-lab Effort on Nuclear Data with a focus on proton induced reactions

Presenter: Dmitri G. Medvedev

Date: January 23, 2023

Co-authors and collaborators

Brookhaven National Laboratory (BNL):

- Michael Skulski (postdoctoral), Cathy S. Cutler

Los Alamos National Laboratory (LANL):

- Ellen O’Brien, Christiaan E. Vermeulen, Eva Birnbaum (alumni), Jonathan Morrell (postdoctoral), Francois M. Nortier (alumni, retired)

Lawrence Berkeley National Laboratory (LBNL)/UC Berkeley:

- Catherine Apgar (graduate student), Morgan Fox (alumni), Andrew Voyles, Lee A. Bernstein

Rationale

Knowledge energy dependent cross section is critical for:

- Accurate prediction of the yield of the isotope of interest
- Accurate prediction of radioisotopic impurities
- Defining the energy windows for optimum, cost-effective production of isotopes

It facilitates:

- Production planning
- Defining shielding requirements
- Transportation and shipment constraints

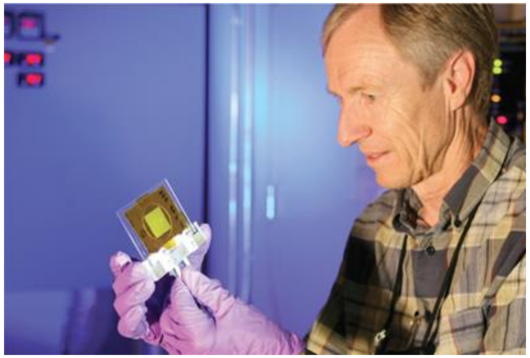
Bonus: comprehensive cross section data sets help constrain nuclear reaction models

Current state of the field

- Most of the data sets are limited to proton energy range up to 30 MeV
- A few more data sets are available up to 160 MeV
- Limited number of data sets are available up to 200 MeV

Approach

A tri-lab collaboration was formed to address the needs and cover proton energy range up to 200 MeV



- LBNL/UC Berkeley – 0 - 55 MeV
- LANL – 55 - 100 MeV
- BNL – 100 - 200 MeV

Seeks to measure nuclear reaction cross sections for production of medically relevant (and other) isotopes and for beam monitor reactions

A comprehensive list of reactions was captured by Dr. François M. Nortier (Meiring) in 2017

Data sets of the primary focus

Priority	Isotope	Target	Incident particle	Measurement focus	Energy range	Years
1	^{72}Se, ^{68}Ge	^{75}As	p	Primary: ^{72}Se, ^{68}Ge Impurities: $^{70,71,73,75}Se$, $^{66,67,69,71}Ge$	Up to 200 MeV	2019
2	^{119}Te, ^{117m}Sn	^{nat}Sb	p	Primary: $^{119m,119g}Te$, ^{117m}Sn Impurities: $^{116,117,118,121m,121g,123m}Te$, $^{113,119m,121m,121g}Sn$	Up to 200 MeV	2020-21
3	^{202}Pb	^{nat}Tl	p	Primary: $^{202m,202g}Pb$ Impurities: $^{198,199,200,201,203,204,205}Pb$	Up to 200 MeV	2022
4	^{134}Ce	^{nat}La	p	Primary: ^{134}Ce Impurities: $^{132,133,133,137,139}Ce$	50-200 MeV	2023

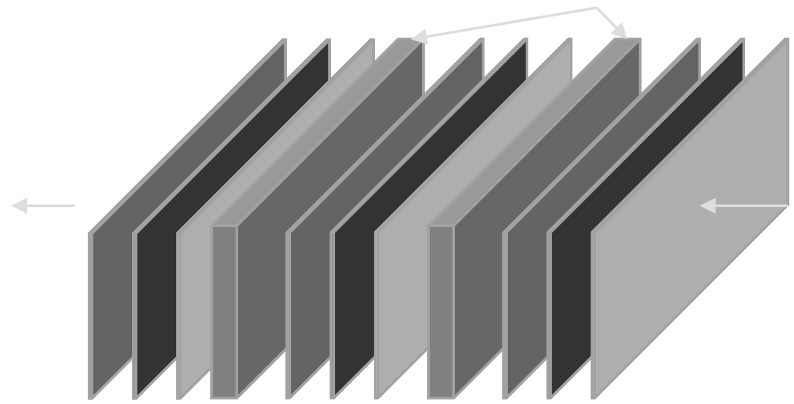
Facilities

- LBNL operates 88-inch cyclotron
  - Both light- and heavy-ion capabilities.
  - Protons and other light-ions are available at intensities (10-20 pA)
  - Maximum energies of 60 MeV (protons), 65 MeV (deuterons), 170 MeV (^{3}He), and 130 MeV (^{4}He).

- BNL (BLIP) and LANL (IPF) share conceptually similar design
  - Operate of proton LINACs
    - IPF max accepted energy 100 MeV, max current 350 μA
    - BLIP max energy 200 MeV, incrementally tunable, max current 200 μA
  - Target stations located 30 feet underground, targets cooled by circulating water
  - Target station cannot be accessed; targets are inserted remotely
  - Designed for large scale isotope production

Experimental: stacked foil technique

- Irradiate array of target and monitor foils with known thickness/areal density intermixed with degraders
  - Very low current 100-200 nA
- Use gamma spectroscopy to count each foil individually
  - Multiple spectra collection allows for accurate error characterization
- Determine activity of all produced radionuclides
- Use activation equation to solve for σ for each individual foil

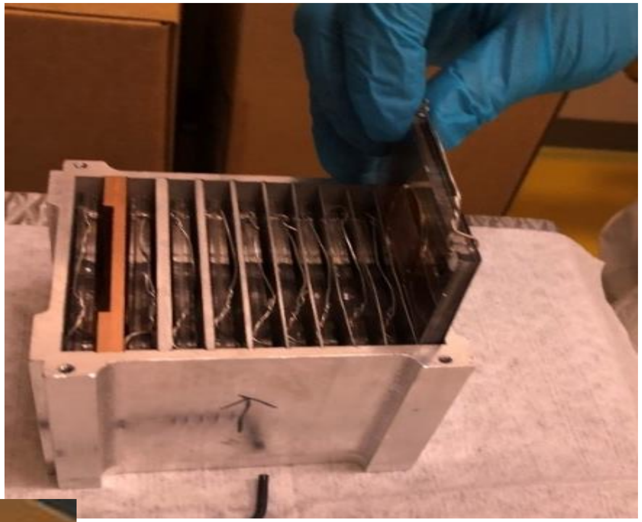


Jon Morrell developed Python based code (Curie)* which performs gamma spectra fitting and decay routine that is used for data analysis

*J. T. Morrell, Curie: Python Toolkit for Experimental Nuclear Data (2020).

Experimental: stacked foils for irradiation

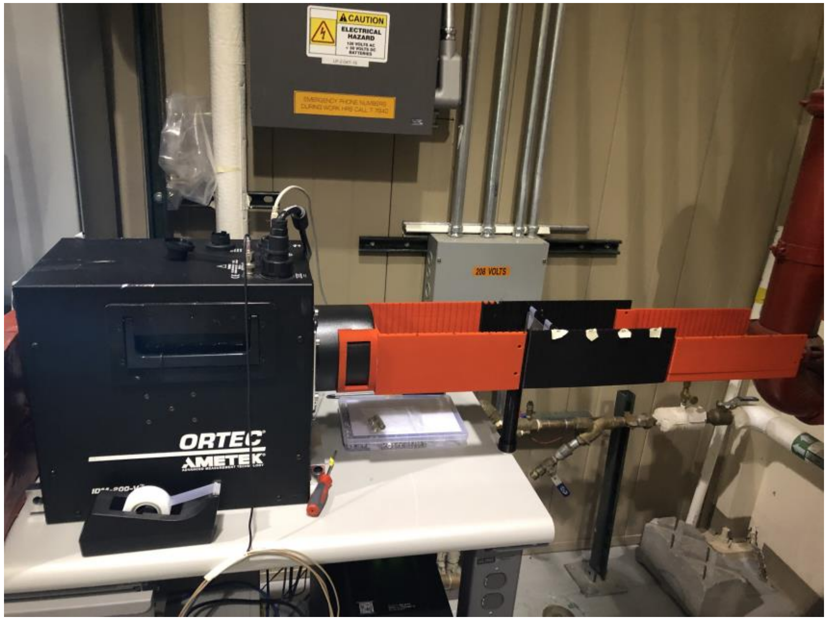
IPF

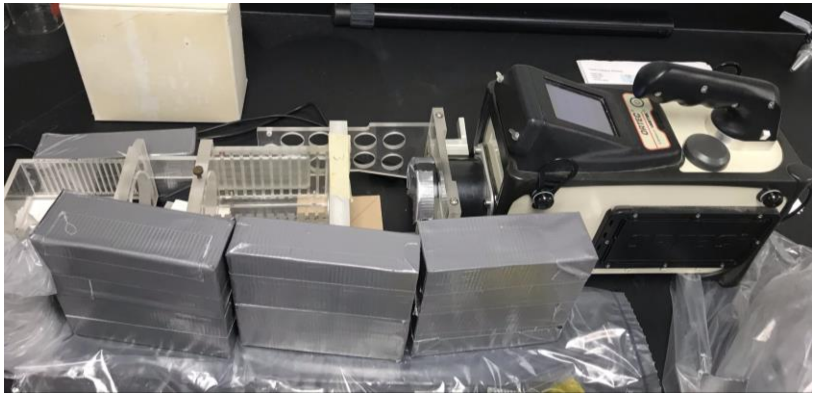


BLIP

LBNL

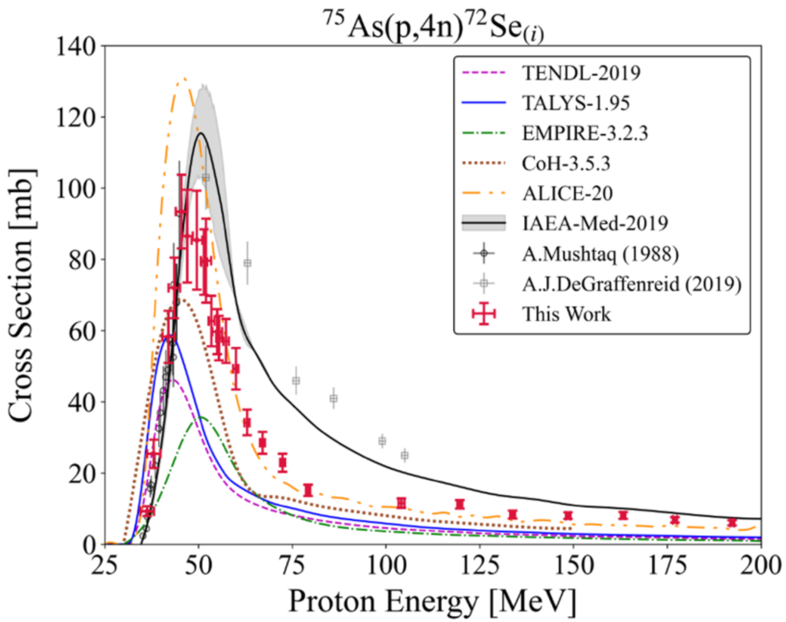
Foil counting set up

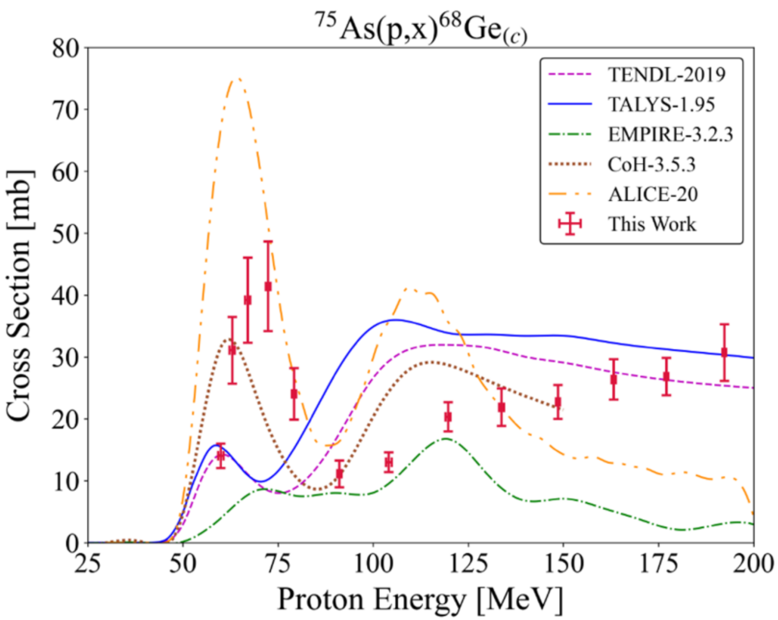




Priority	Isotope	Target	Incident particle	Measurement focus	Energy range	Years
1	^{72}Se, ^{68}Ge	^{75}As	p	Primary: ^{72}Se, ^{68}Ge Impurities: $^{70,71,73,75}Se$, $^{66,67,69,71}Ge$	Up to 200 MeV	2019

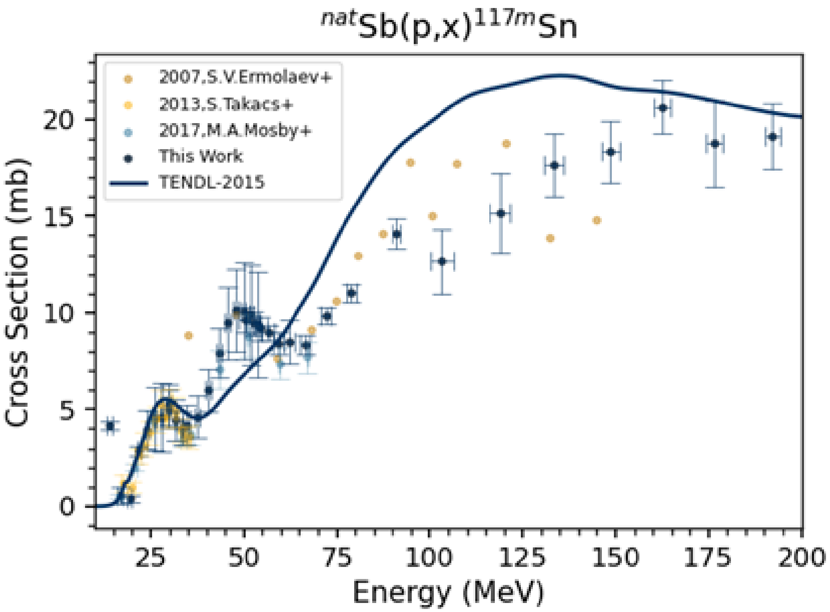
Morgan Fox
LBNL

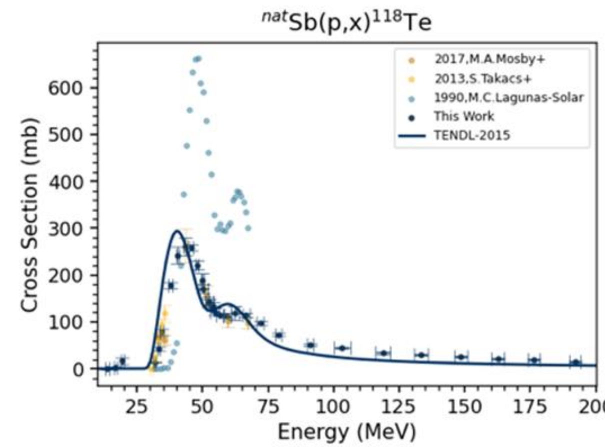


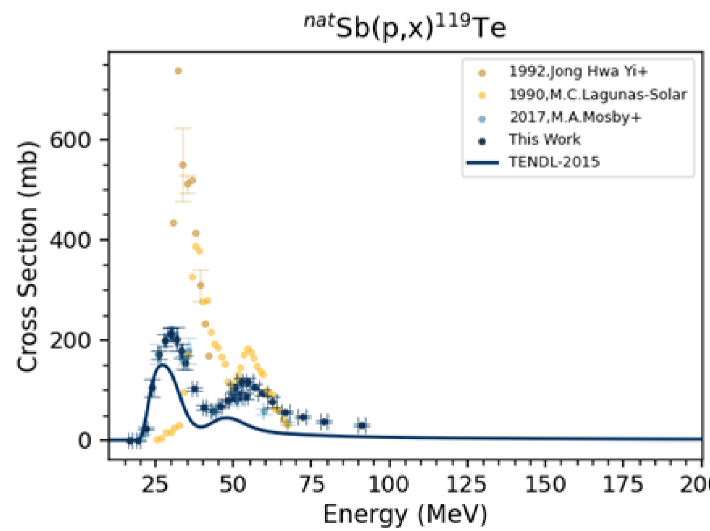


Priority	Isotope	Target	Incident particle	Measurement focus	Energy range	Years
2	^{119}Te, ^{117m}Sn	^{nat}Sb	p	Primary: $^{119m,119g}Te$, ^{117m}Sn Impurities: $^{116,117,118,121m,121g,123m}Te$, $^{113,119m,121m,121g}Sn$	Up to 200 MeV	2020-21

Catherine Apgar
LBNL



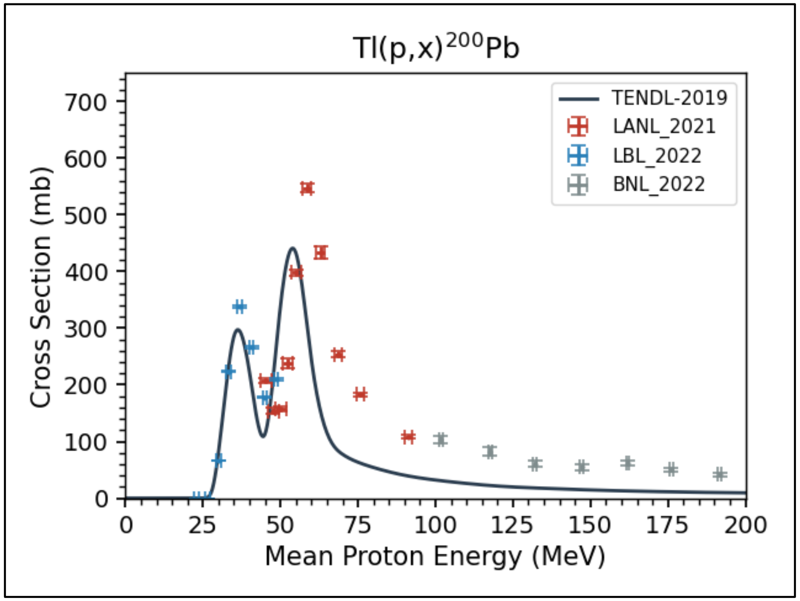


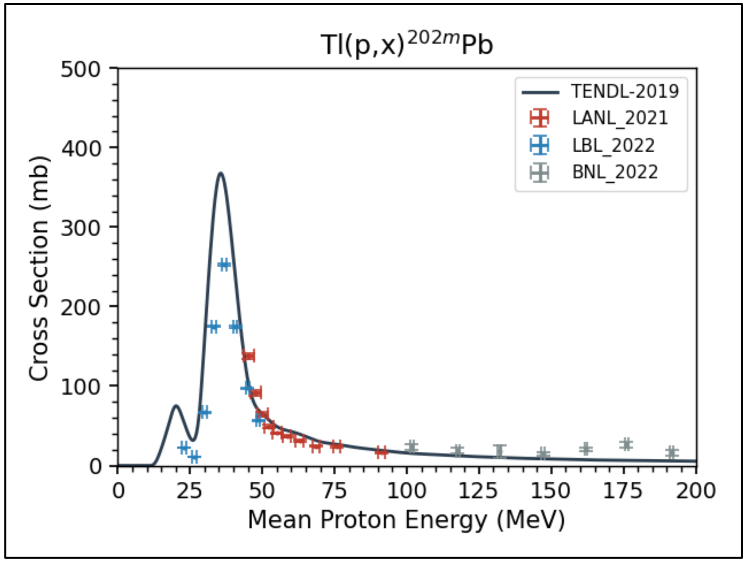


Brookhaven National Laboratory

Priority	Isotope	Target	Incident particle	Measurement focus	Energy range	Years
3	^{202}Pb	^{nat}Tl	p	Primary: ^{202m,202g}Pb Impurities: ^{198,199,200,201,203,204,205}Pb	Up to 200 MeV	2022

Michael Skulski
BNL





Priority	Isotope	Target	Incident particle	Measurement focus	Energy range	Years
4	^{134}Ce	^{nat}La	p	Primary: ^{134}Ce Impurities: ^{132,133,133,137,139}Ce	50-200 MeV	2023

Jonathan Morrell
LBNL→LANL

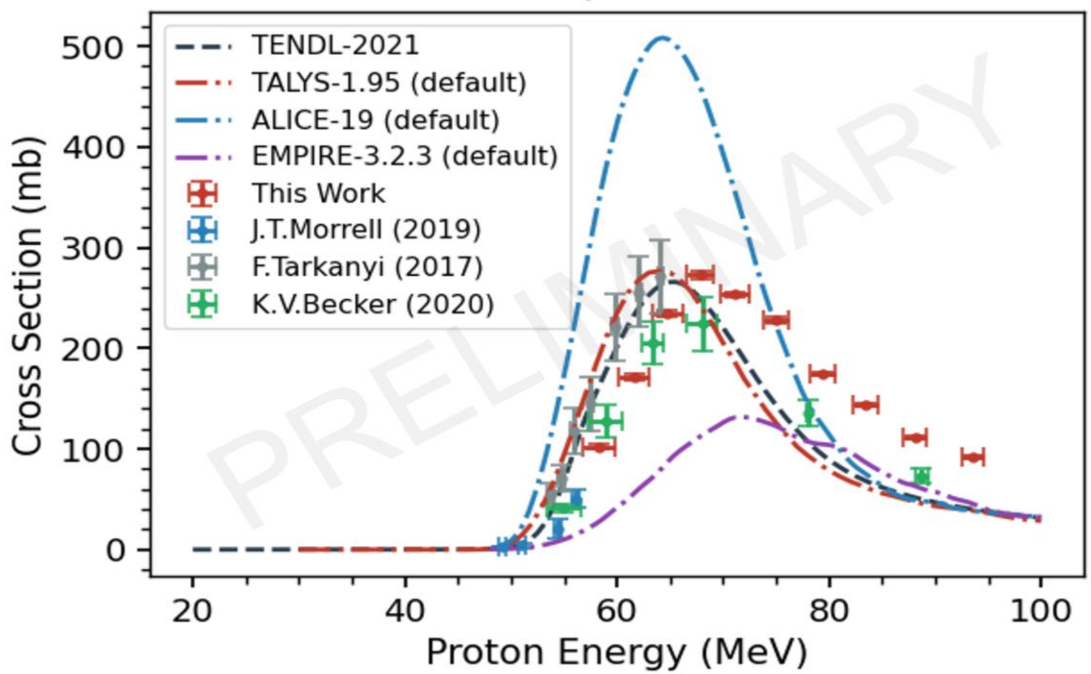


Figure 1. Preliminary measurement of the ^{139}La(p,x)^{134}Ce reaction cross section, from the Sep. 22 experiment at LANL IPF, plotted against existing measurements and several modeling codes (using default parameters).

Project Impact so far

- More than 55 datasets for As(p,x) and monitor reactions were collected and analyzed
- Se-72, Se-73, Se-75 data sets were extended to 200 MeV
- First data set for As(p,x) ^{68}Ge reaction up to 200 MeV
- Improved predictive power of Talys
- Workforce training component:
  - Jonathan Morrell (UC Berkeley), now postdoctoral scholar at LANL
  - Morgan Fox (UC Berkeley), accepted position in Canada
  - Catherine Apgar (currently graduate student at UC Berkeley) Michael Skulski – postdoctoral scholar at BNL

Morgan B. Fox, Andrew S. Voyles, Jonathan T. Morrell, et al. Investigating high-energy proton-induced reactions on spherical nuclei: Implications for the preequilibrium exciton model. *Physical Review C*, 103, pp. 034601 (2021

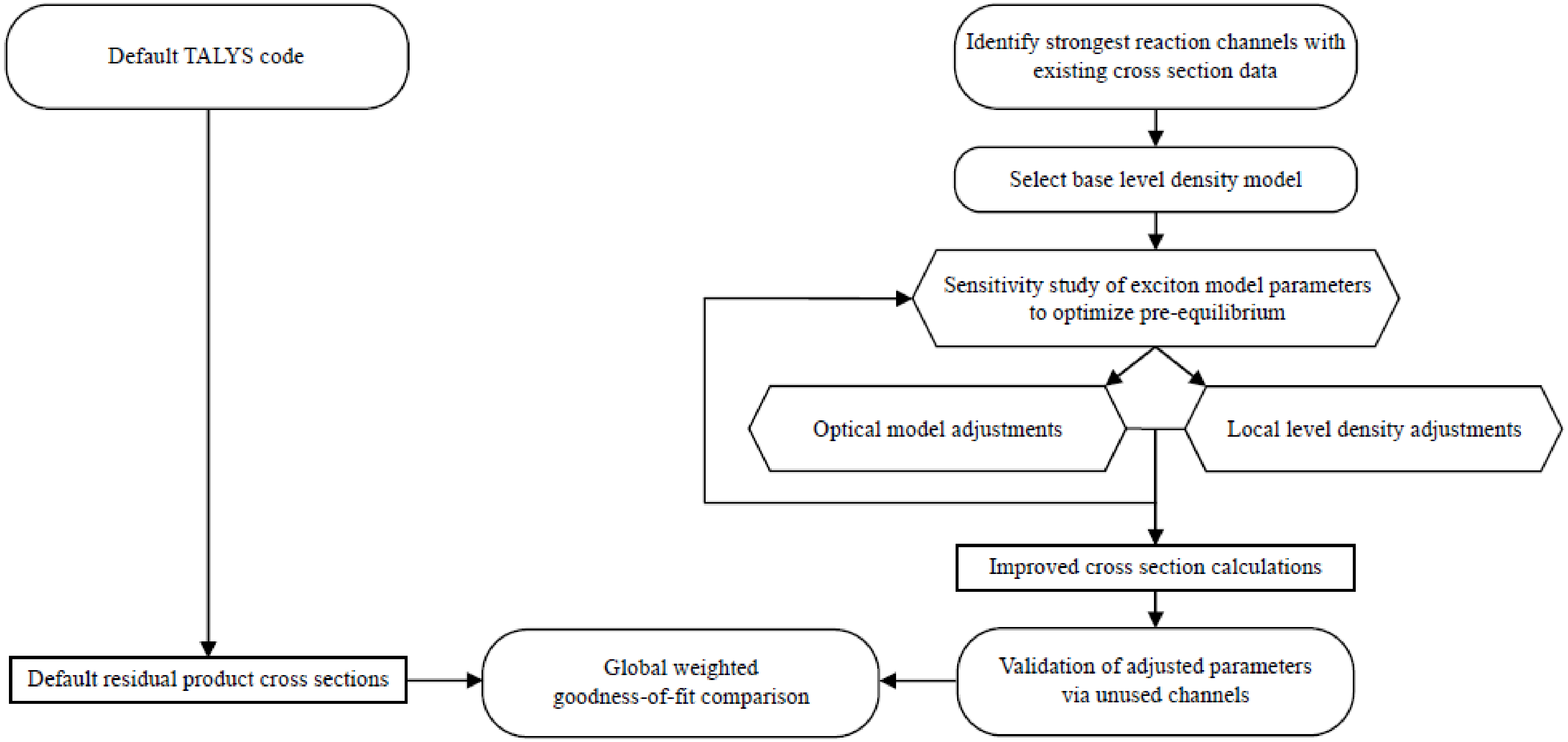
Morgan B. Fox, Andrew S. Voyles, Jonathan T. Morrell, et al. Measurement and Modeling of Proton-Induced Reactions on Arsenic from 35 to 200 MeV, submitted June 2021

Future work

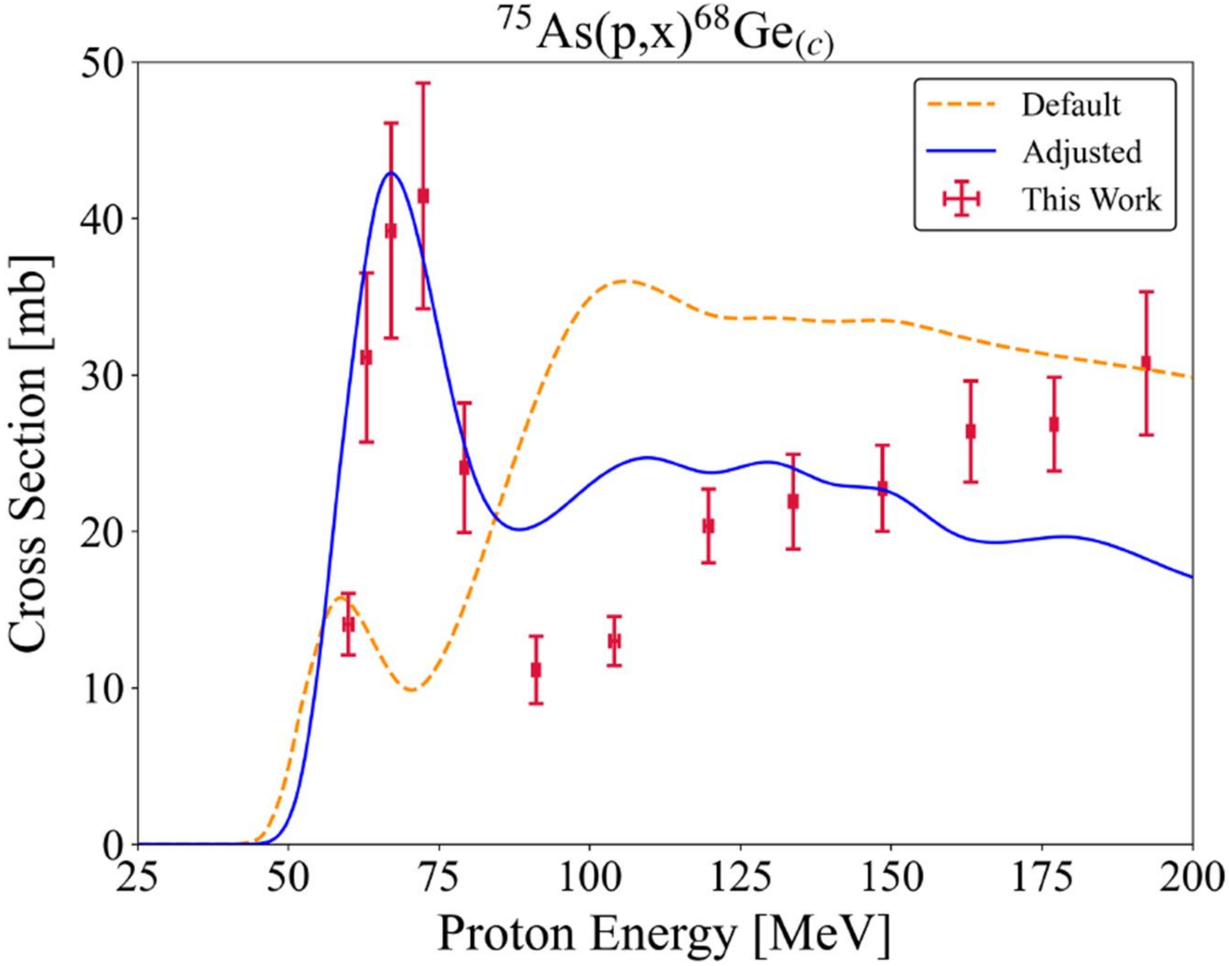
Development of a Machine Learning-Augmented Reaction Evaluation and an Accessible End-User Interface for Radioisotope Production

Talys nuclear reaction code improvement

Morgan Fox
LBNL



Comparison of default and adjusted Talys calculations



Morgan Fox
LBNL

Work scope for the coming years

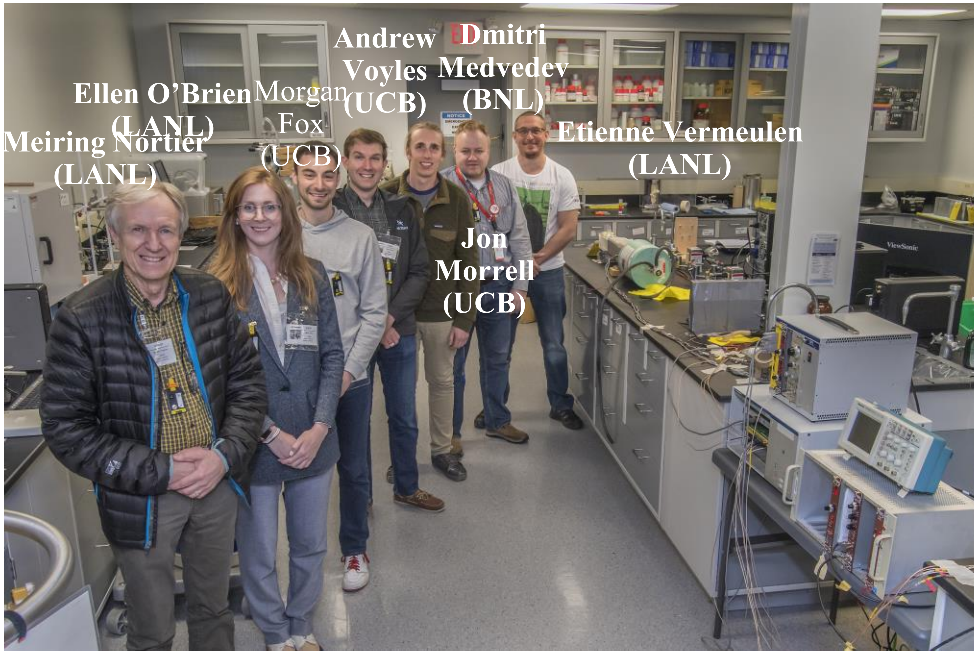
- AI/ML powered algorithm training using existing TREND measurements
- Additional high-energy cross section measurements to bolster the nuclear physics model performance
- Improving existing recommended cross section database and integrating into a user-friendly yield calculator

Target	Isotope(s) of interest and application
^{nat}Ag	^{103}Pd (brachytherapy)
^{169}Tm	^{161}Tb (radioimmunotherapy)
^{89}Y	^{88}Zr (^{88}Zr/^{88}Y generator)
^{209}Bi	^{203}Pb (SPECT imaging)
^{nat}Ir	^{188}Pt (Auger emitter)
^{232}Th	^{225}Ac, ^{225}Ra (radioimmunotherapy)
^{nat}Gd	^{152}Tb (PET Imaging)
^{45}Sc	^{44}Ti (^{44}Ti/^{44}Sc generator)
^{197}Au	^{188}Pt (Auger emitter)

Acknowledgement

- US Department of Energy Isotope Program for funding
- Whole-hearted acknowledgement goes to all support personnel of LINAC, Health Physics, engineers, designers
- BNL is managed by Brookhaven Science Associates operated by Battelle Memorial Institute (Battelle) and Stony Brook University for US DOE Office of Science.
- LANL is managed by Triad LLC, a partnership between Battelle, the Texas A&M University System, and the Regents of the University of California (UC) for the Department of Energy’s National Nuclear Security Administration.
- LBNL is managed by University of California for US DOE Office of Science

The Tri-lab Nuclear Data Collaboration



Not Pictured:

Lee Bernstein (LBNL)

Eva Birnbaum (LANL)

Cathy Cutler (BNL)