Hauser-Feshbach Analysis of Fast Neutron-Induced Reactions on Chlorine

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Motivation

Neutron-induced reactions on chlorine isotopes are critical channels for many modern applications. In the astrophysical *s*-process, ^{35}Cl(n,p)^{35}S plays an important role in isotopic generation in its mass region, particularly of the rare ^{36}S [1-3]. In nuclear energy, this same cross section spans the crucial energy range for advanced molten salt fast fission reactor designs [4], while similar channels on ^{37}Cl dictate the design of tritium breeding blankets for fusion systems [5]. Such cross sections have been measured recently at the Los Alamos Neutron Science Center (LANSCE) in partnership with TerraPower LLC (TP) for application to their Molten Chloride Reactor Experiment (MCRE) [6]. These data have enabled a new Hauser-Feshbach analysis, also led by LANL, of the "fast" (statistical) energy region crucial for the MCRE and other applications, yet previously not well constrained. In general, a reduction in neutron absorption into charged particle channels for this mass region has been observed; however, challenges persist for the particular case of ^{35}Cl due to its prominent structural deficiencies.

Statistical (Fast-Energy) Analysis

With the combination of new data from LANSCE and that available in the literature, a statistical re-analysis was performed on the (n + ^{35}Cl) system on all relevant channels. The code CoH$_3$ [7] was utilized, allowing inclusion of coupled-channels effects and basic nuclear structure models into the standard Hauser-Feshbach and pre-equilibrium treatments. The results of this analysis for the total and (n,p) cross sections on ^{35}Cl (the primary target) and natural extensions to neighboring ^{37}Cl and ^{39}K are shown in **Figure 2**.

(T. Kawano [7])
Hauser-Feshbach + coupled channels & basic nuclear structure models

Key findings of the analysis are:

- a necessary **reduction in neutron absorption** through primarily the (n,p) channel, across multiple isotopes;
- strong **fluctuations in the ^{35}Cl(n,p) cross section** that deviate significantly from the statistical trends;
- large (~50%) **increases to the single-particle pre-equilibrium** emission components at higher energies (>10 MeV).

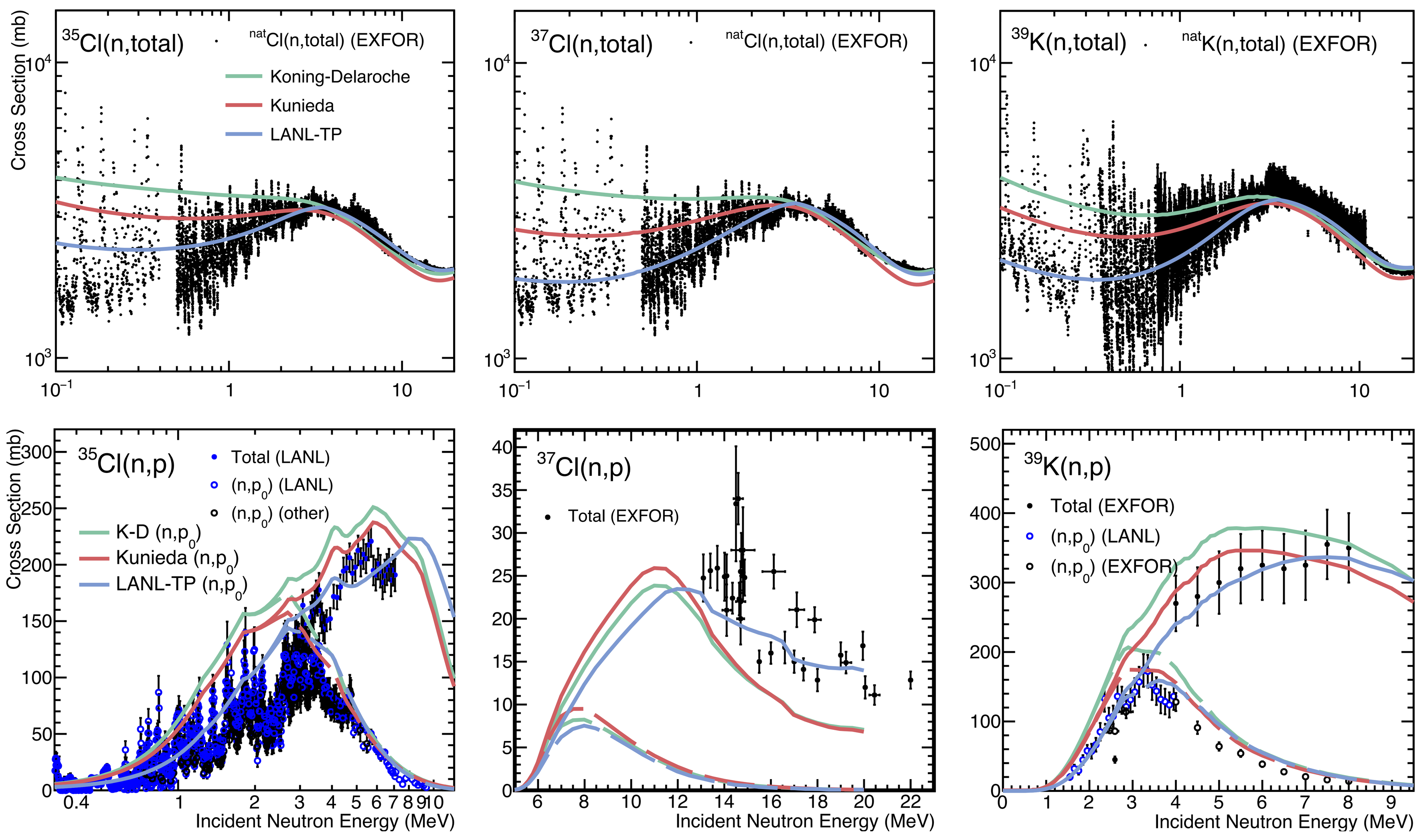
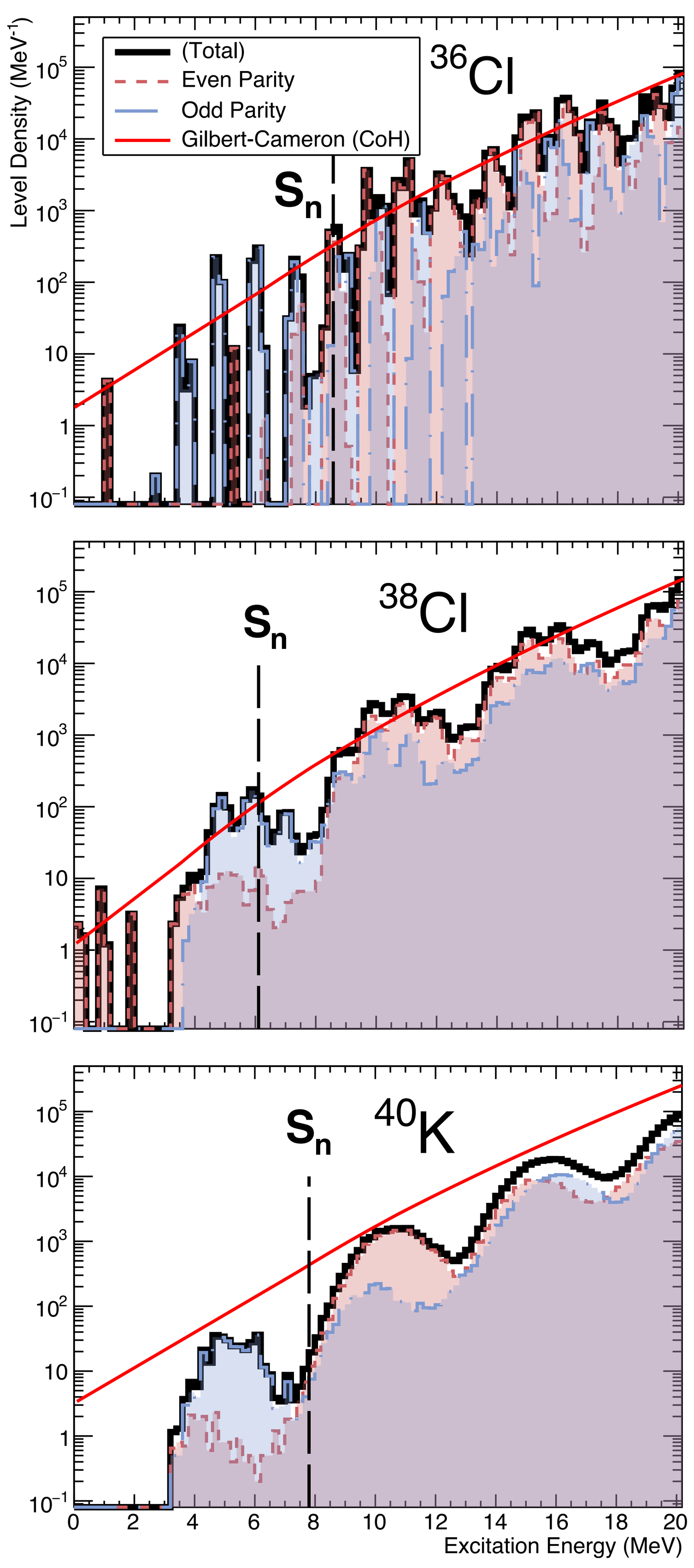


Figure 2 (above) Statistical analyses of select reactions on ^{35}Cl and related isotopes. Compared against the base calculations using the global optical models of Koning & Delaroche [8] and Kunieda *et al.* [9] are this work's results fine-tuning to the (n + ^{35}Cl) channels, in which modifications were made to the Kunieda OMP, Gilbert-Cameron-parameterized level densities, and pre-equilibrium state densities of the two-component exciton model. Note that the reduction in neutron absorption through (n,p) found necessary for ^{35}Cl also seems to apply to ^{39}K, for reasons discussed in the section below.



Structural Considerations

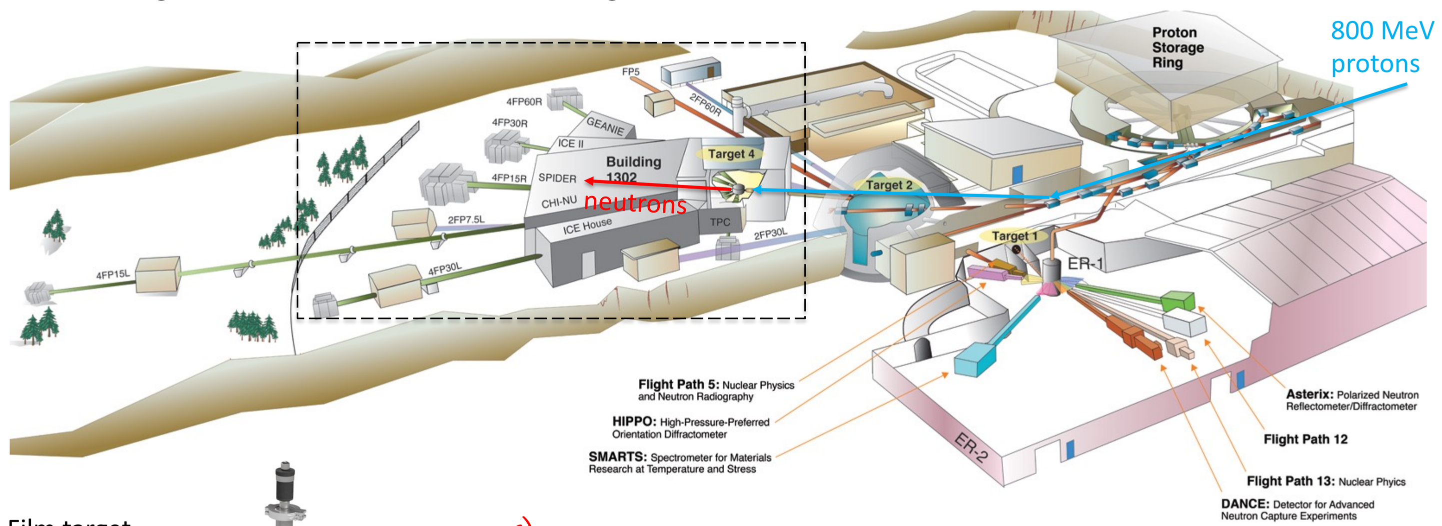
The Hauser-Feshbach model assumes a smooth and dense trend of compound structural levels with excitation energy. However, ^{36}Cl (the compound of n + ^{35}Cl) appears not to follow this trend. **Figure 3** shows a set of combinatorial level density calculations for the relevant compound nuclei ^{36}Cl, ^{38}Cl, and ^{40}K using the Finite Range Droplet Model (FRDM). Compared as the solid red lines are the H-F level densities calculated by CoH under the Gilbert-Cameron (G-C) parameterization [10].

Proximity to the Z=N=20 shell closure encourages a deviation (deficiency) from the purely statistical predictions, across all three isotopes. Thus the reduction in absorption found through the analysis on ^{35}Cl applies locally as well. However, as mass (primarily neutron number) is decreased toward ^{36}Cl, single-particle occupancies across the neutron Fermi surface become less "smooth" in transition, leading to more sporadic structure. ^{36}Cl in particular demonstrates several short-range gaps and jumps around the neutron separation energy. Therefore formation of the compound is hindered for ~few MeV incident neutron energies, and the cross section is prone to fluctuations beyond the Hauser-Feshbach predictions, as seen above.

Figure 3 (left) FRDM combinatorial calculations for the level densities of relevant chlorine and potassium isotopes. Contrasted against the total (black) are the individual components for even (dashed red) and odd (dashed blue) parities. Configurations up to 5p-5h are included. Also noted are the neutron separation energies S$_n$ and the Gilbert-Cameron (H-F) level densities calculated by CoH (dashed-black verticals and solid red curves respectively).

Experimental Methods

Absolute cross sections for (n,p) and (n,α) reactions on ^{35}Cl, ^{39}K, and ^{40}K have been measured at the LANSCE white neutron source over several run cycles [11-13]. The data were taken using the Low-Energy (n,Z) instrument (LENZ) at the Weapons Neutron Research facility (WNR) (**Figure 1**), where the 800 MeV proton beam from the linear accelerator is impinged on an unmoderated tungsten spallation target. The film targets in the LENZ chamber exposed to the resulting white neutron fluence were ^{35}Cl-enriched AgCl and NaCl, and KCl enriched to 13% in ^{40}K, on gold or brass backings. The use of two different neutron flight paths relative to the spallation target allowed a coverage of incident neutron energies from 300 keV – 12 MeV.



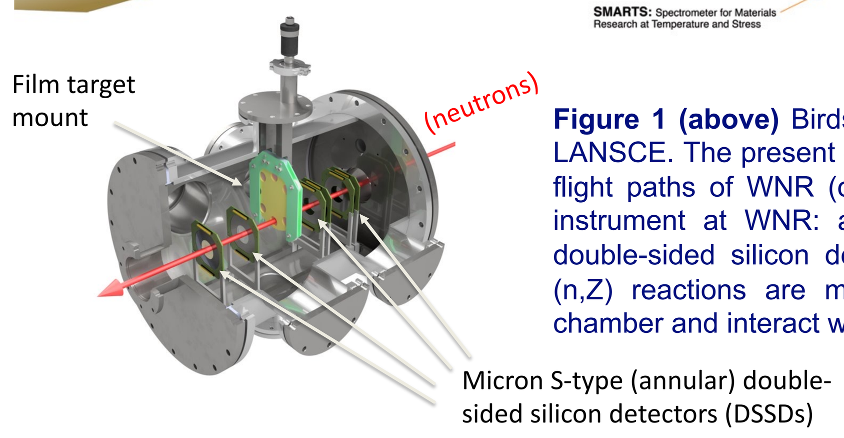


Figure 1 (above) Birds-eye view of the WNR and Lujan facilities at LANSCE. The present experiments were conducted at the 15R & 90L flight paths of WNR (outlined in the dashed box). **(left)** The LENZ instrument at WNR: an aluminum vacuum chamber with several double-sided silicon detectors (DSSDs) around a solid film target. (n,Z) reactions are measured as neutrons (n) pass through the chamber and interact with the target to create charged particles (Z).

Impact & Conclusions

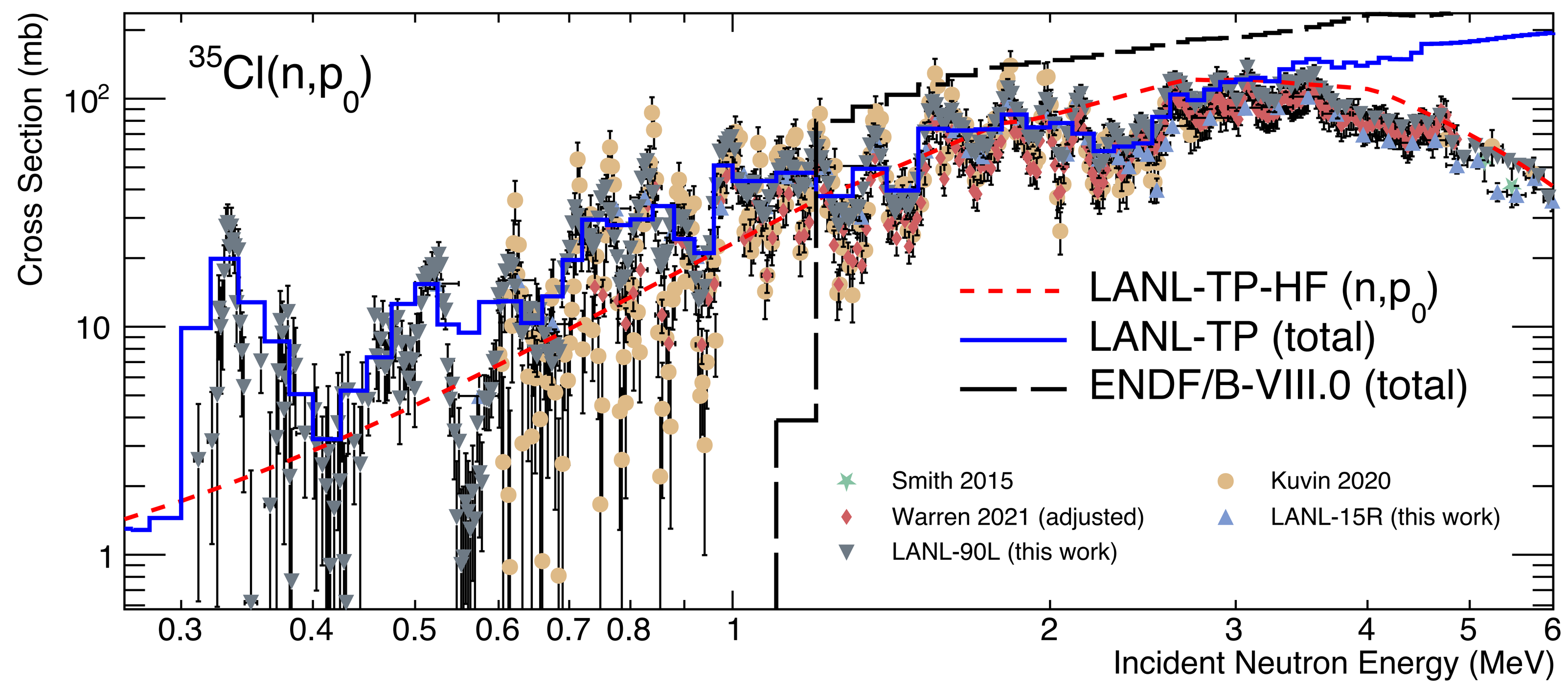


Figure 4 Solution to the structurally-based fluctuations in the ^{35}Cl(n,p) cross section for the *evaluated* data used in applications. The purely statistical calculation (LANL-TP-HF) has been used as a prior for a direct but coarse fitting of the experimental data (LANL-TP). Shown for comparison is the current version in ENDF/B-VIII.0, where the statistical component is cut off abruptly at 1.2 MeV to drop several orders of magnitude.

>> To account for the average effect of the fluctuations in the ^{35}Cl(n,p$_0$) cross section, the data have been fit coarsely before being formatted into an ENDF-6 file for application and validation.

>> Preliminary calculations by TerraPower with the MCRE indicate a net increase in reactivity by ~60% compared to previous evaluations, due to the reduced neutron absorption.

>> In the temperature region important for the *s*-process (kT = 25-200 keV), this work's newly constrained Maxwellian-Averaged Cross Section (MACS) for ^{35}Cl(n,p) lies between previous (unconstrained) evaluations.

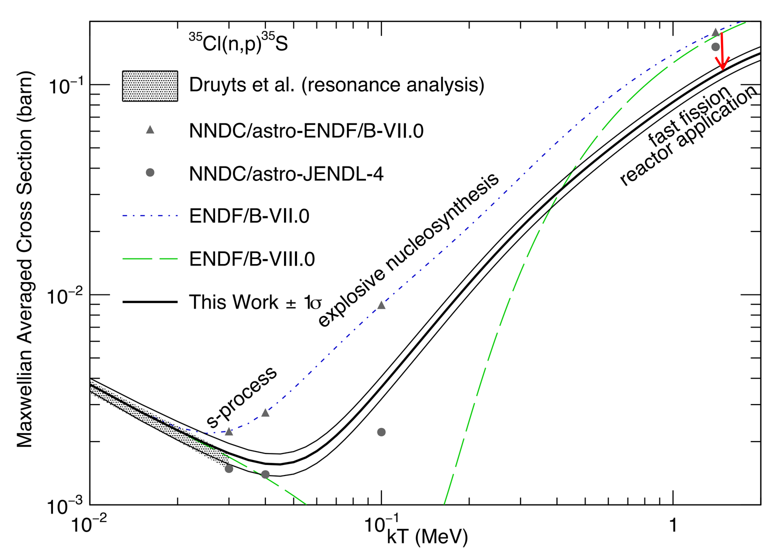


Figure 5 Maxwellian-Averaged Cross Sections (MACS) for ^{35}Cl(n,p), comparing this work's results with those of currently available evaluations.

Future Work

- These calculations form the foundation for a suite of improved evaluations performed by LANL for use by the nuclear data community.
- The ^{35}Cl evaluation is already available on the ENDF GitLab for download (contact K.H. for details).
- New fast-energy data for ^{37}Cl are necessary to improve its model description; such measurements are being considered for a future LANSCE run cycle.
- Preliminary new data for ^{35}Cl in the resonance region down to thermal have been taken at LANSCE's Lujan Center, for eventual low-energy extension of the re-evaluation (funded by NCSP).

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