Nonstatistical fluctuations in the $^{35}Cl(n, p)^{35}S$ reaction cross section at fast-neutron energies

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Informing the design of next-gen reactors

- Next-generation reactors are intended to be safer, more reliable, more sustainable, and beneficial to non-proliferation.
- Various designs are under consideration/development, including Fast Spectrum Molten Salt Reactors.
- The use of chloride salts, as an alternative to fluoride salts, comes with a variety of benefits, but significant uncertainties have delayed their exploration.
- Chlorine has two stable isotopes: ^{35}Cl(76%) and ^{37}Cl(24%)
- The dominant reaction in a fast spectrum molten salt reactor, using chloride salts, is the $^{35}Cl(n,p)^{35}S$ reaction. (^{35}S $T_{1/2}$ ~ 75 days)

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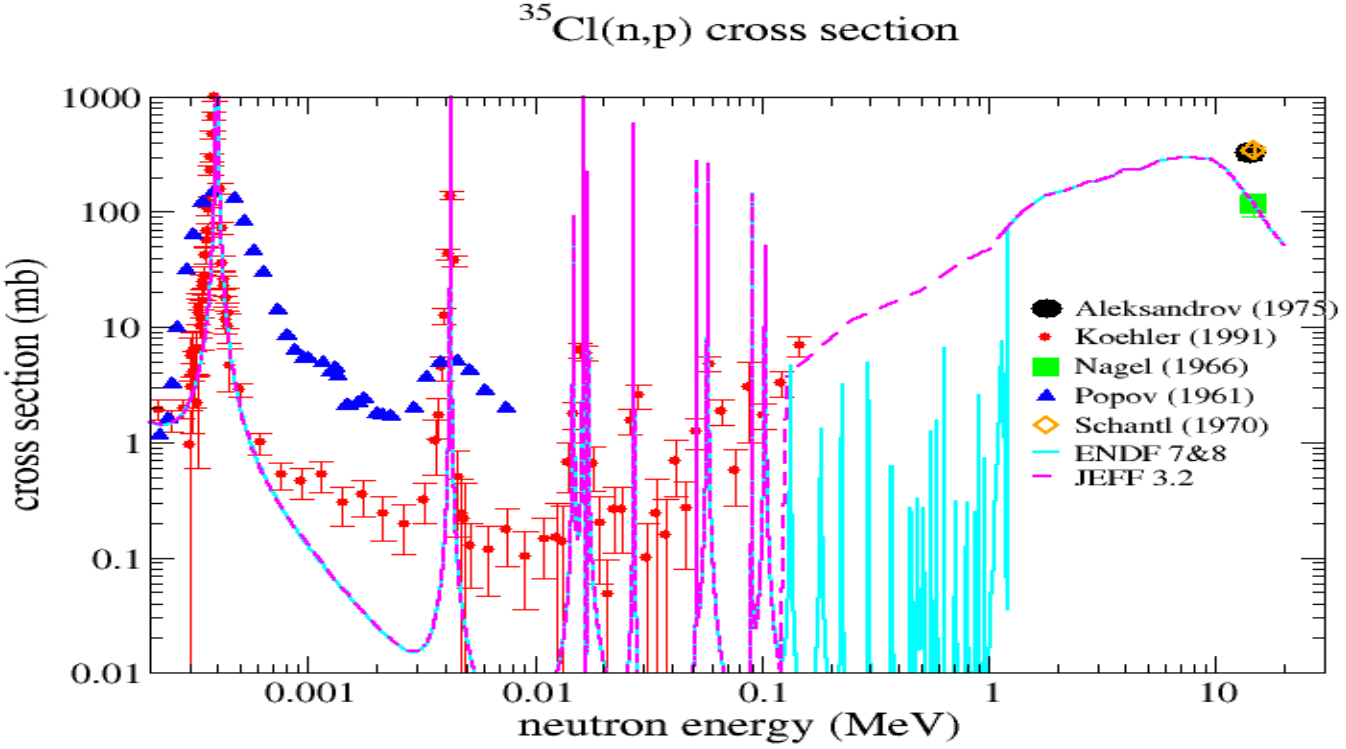
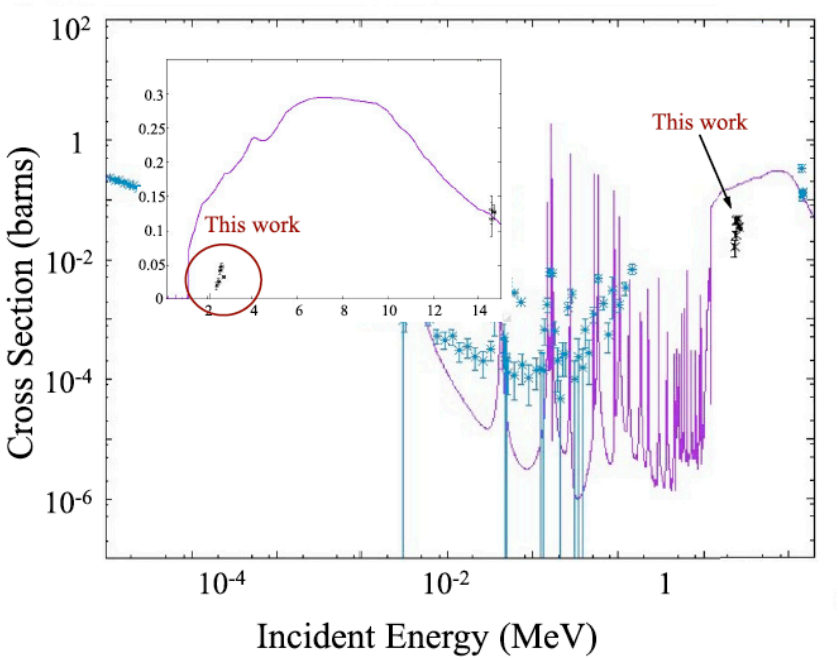
- Studies have shown that the change in the $^{35}Cl(n,p)$ cross-section evaluation between different versions of ENDF-VII lead to significant changes in the reactivity of the system (>5000 pcm) http://www.oecd-nea.org/dbdata/hprl/

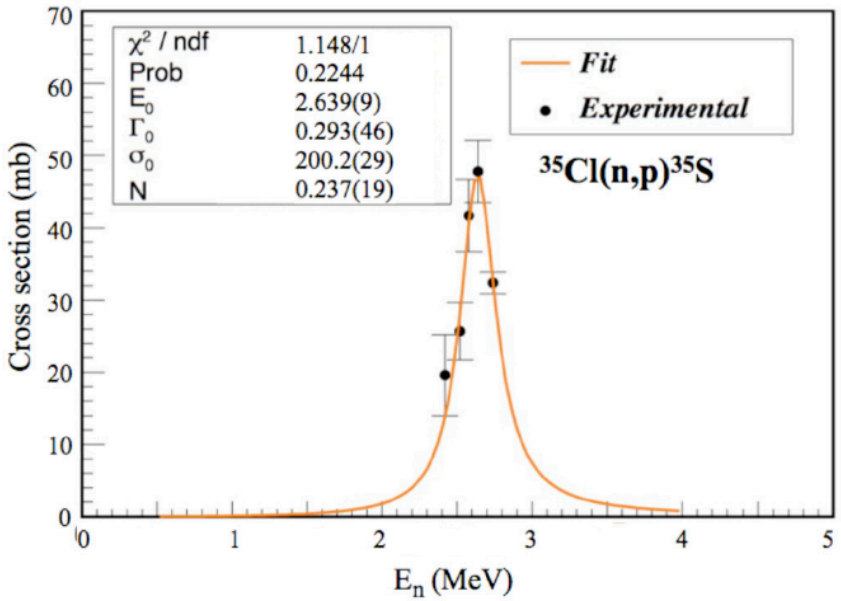


Evidence of nonstatistical properties at ~2.6 MeV

- Recent measurement of ^{35}Cl(n,p) and ^{35}Cl(n,a) at incident neutron energies between 2.42 and 2.74 MeV shows a hint of nonstatistical behavior and a reduced cross-section relative to all data libraries.

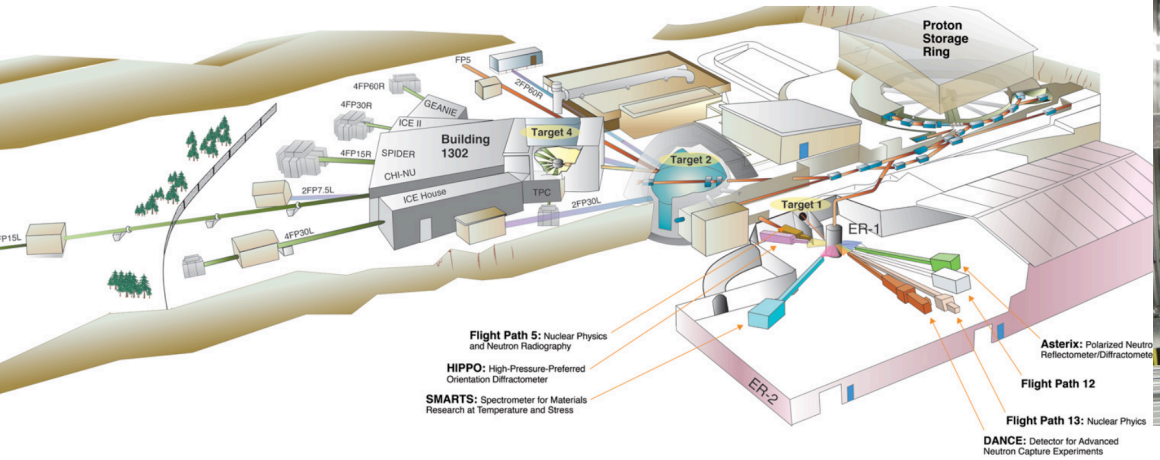
Batchelder et al. PRC99,044612(2019)

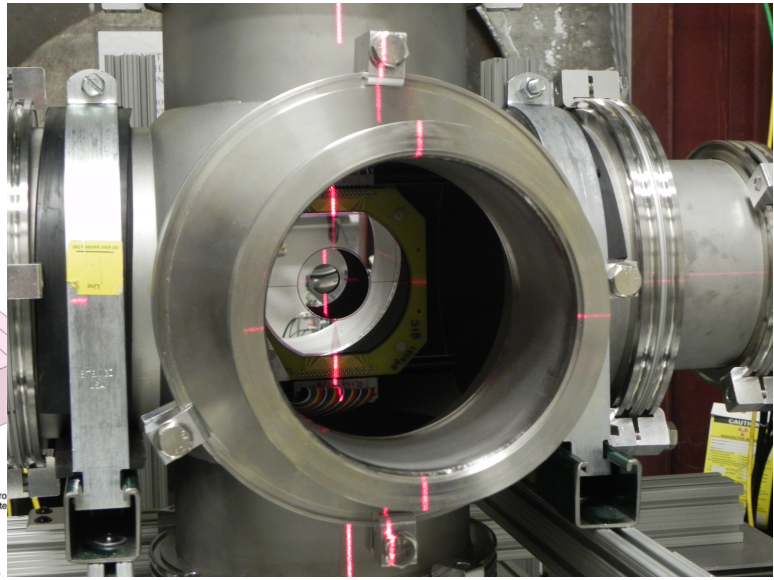




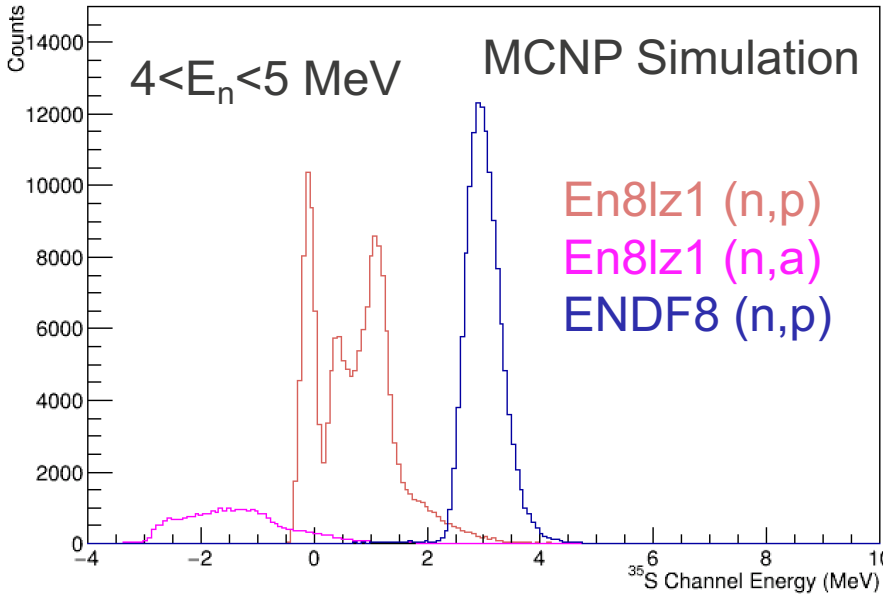
Experimental Setup

- WNR Facility at LANSCE: fast neutrons with a broad energy spectrum ~100s of keV to ~100s of MeV
- Annular silicon detectors for detecting charged particles
- 350 and 450 ug/cm2 NaCl (^{35}Cl enriched) targets evaporated on to 6um thick brass foils
- Two flight paths: 15R (15.2m, forward angles) 15R(14.1m, backward angles) and 90L (8.1m, backward angles)



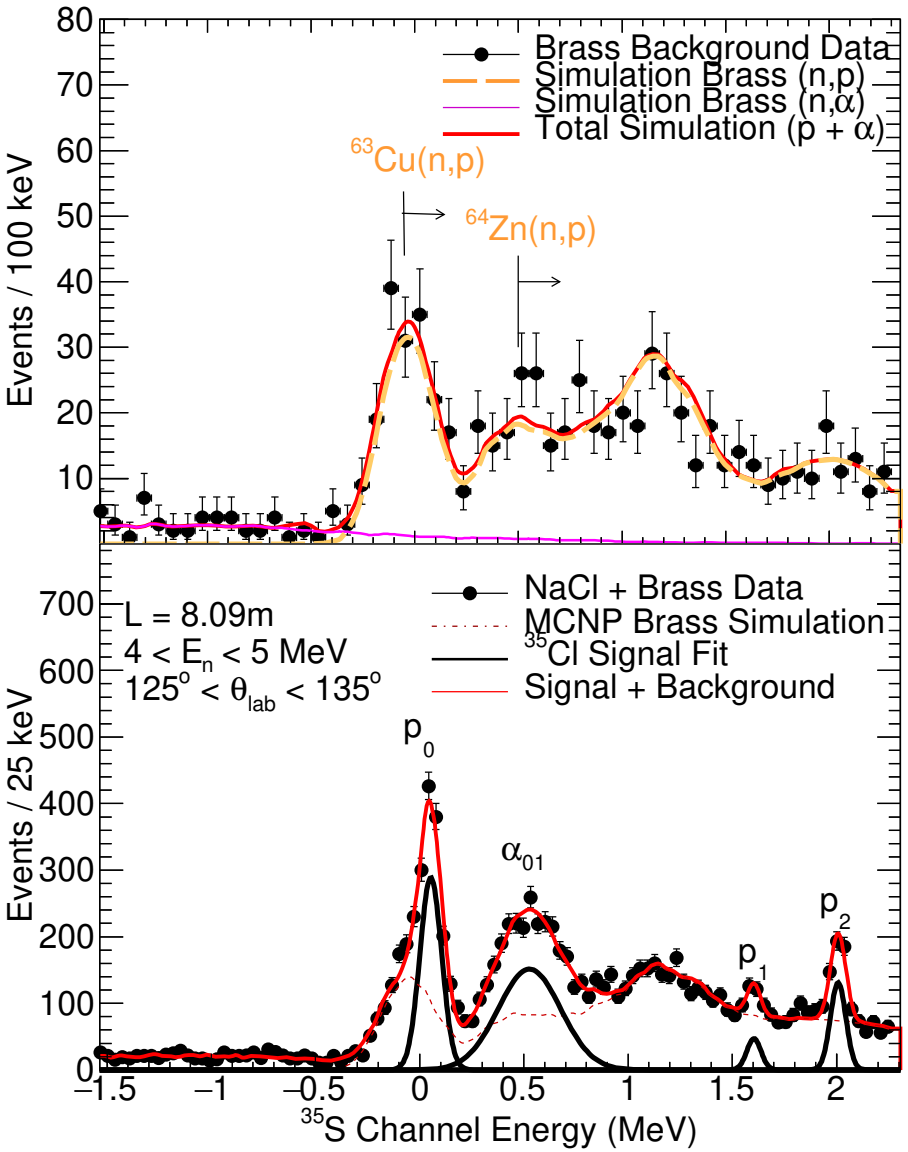


Measurement of $^{35}Cl(n,p)^{35}S$



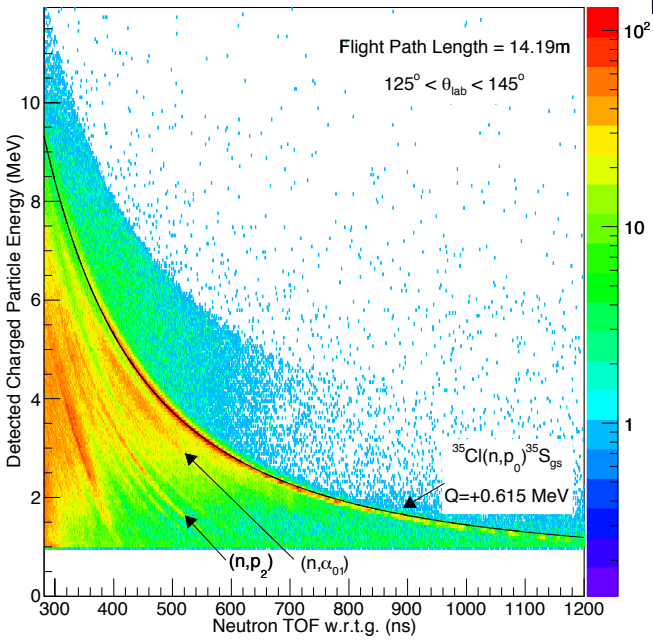
- MCNP simulations using the modified evaluation (H.I. Kim's talk) built on ENDF8 provides good reproduction of the experimental spectra.

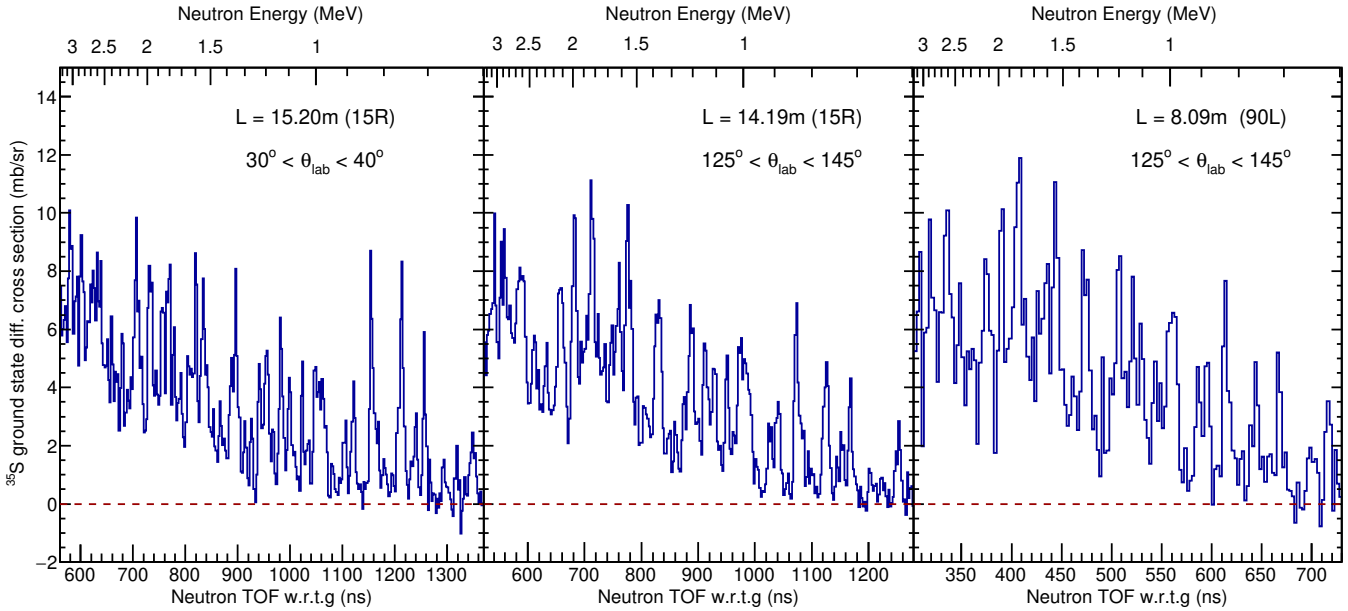
- Signals from $^{35}Cl(n,p)$ and $^{35}Cl(n,a)$ are observed on top of the brass background.



Measurement of $^{35}Cl(n,p_0)^{35}S_{gs}$

- Non-statistical fluctuations are observed in the $^{35}Cl(n,p_0)^{35}S_{gs}$ cross section extending up to ~3 MeV, consistent with the observation by Batchelder *et al.*





$^{35}Cl(n,p)^{35}S$ Results

- Consistent with the results of Batchelder *et al.* the $^{35}Cl(n,p)$ cross-section is over-predicted by ENDF/B-VIII.0 at energies near ~3 MeV.

- Our results for $^{35}Cl(n,p_0)$ between 4 and 6 MeV are in good agreement with the measurement of Smith *et al.* who inferred the cross section based on measured CLYC detector yields (efficiency).

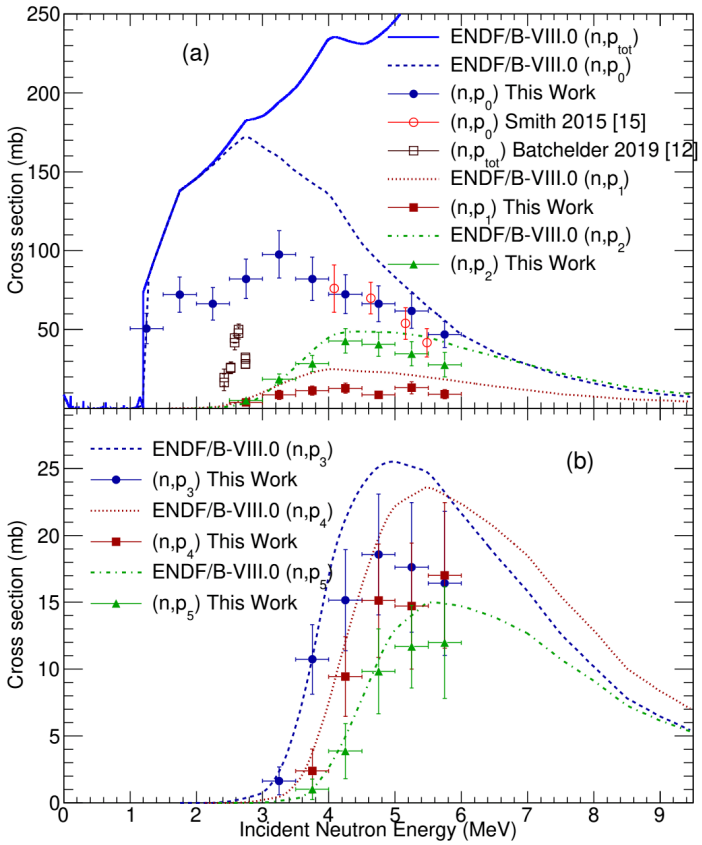


FIG. 11. Partial angle-integrated cross sections in 500 keV wide bins are shown in comparison to the ENDF/B-VIII.0 evaluated partial cross sections for (n,p) reactions up to (a) the first 2 excited states of ^{35}S and (b) up to (n,p_5) The results in this work show a significant reduction relative to the ENDF/B-VIII.0 cross section for the (n,p_0) reaction channel between 1 and 5 MeV and a slight reduction for the excited states.

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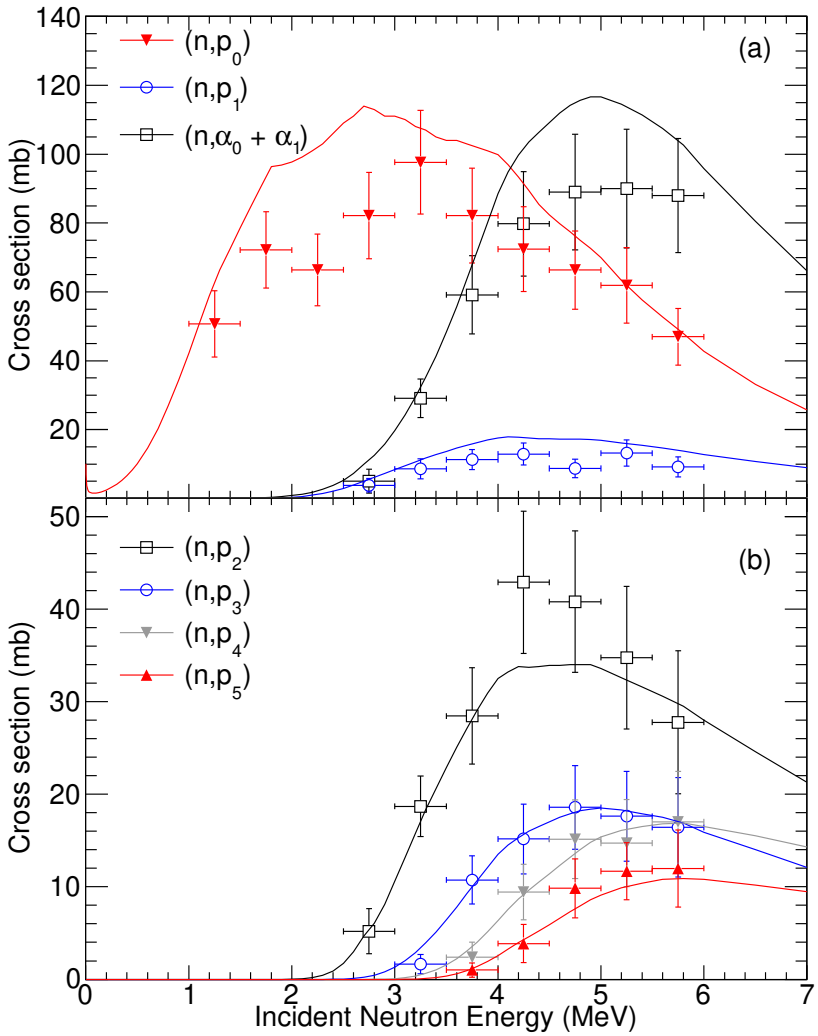
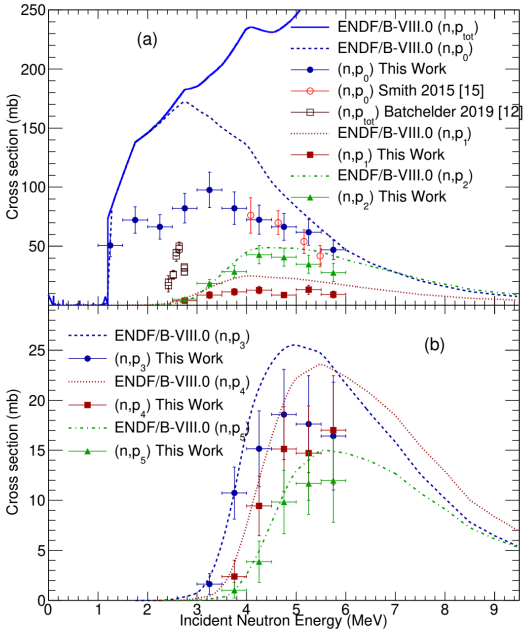
- Our results for $^{35}Cl(n,p_0)$ between 4 and 6 MeV are in good agreement with the measurement of Smith *et al.* who inferred the cross section based on measured CLYC detector yields (efficiency).
- The energy dependent trends could be reproduced reasonably-well using a statistical Hauser-Feshbach calculation (CoH3) using a modified Kunieda potential. (T. Kawano from T-2)



states.

$^{35}Cl(n,p)^{35}S$ Results

- LENZ data indicates that the ENDF/B-VIII.0 evaluation of the $^{35}Cl(n,p)^{35}S$ reaction overestimates the cross section above 1.25 MeV and dramatically underestimates it below 1.25 MeV. A full re-evaluation is recommended.



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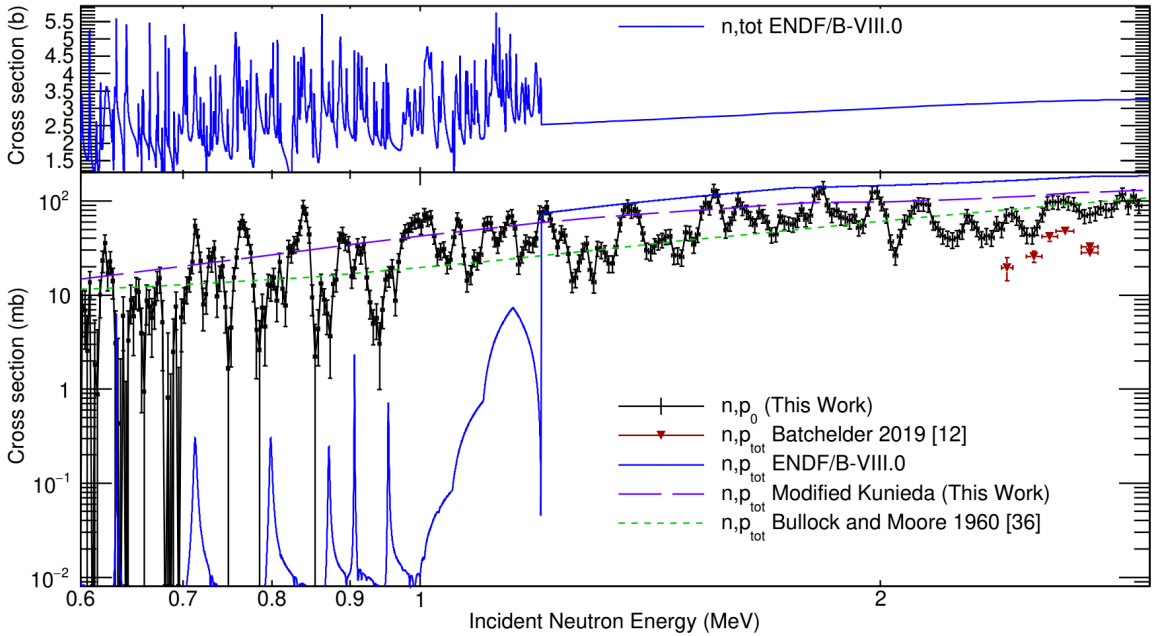

IG. 13. Bottom: The partial angle-integrated cross section for populating the ground state of ^{35}S. The data is reasonably ell bounded, albeit with significant fluctuations, by the calculated results adopted from Ref. [36], and from the statistical alculation employing a modified Kunieda potential from this work. For comparison, the top panel shows the resonance tructures in the ^{35}Cl(n,total) spectrum of ENDF/B-VIII.0. The resonances that we observe in the (n,p) data are clearly the um of multiple narrower resonances that appear to have analogues in the (n,total) data but are not strongly represented in he (n,p) evaluation below 1 MeV.

Summary/Outlook

- Definitively confirm the non-statistical behavior of the $^{35}Cl(n,p)^{35}S$ reaction up to and around ~3 MeV.
- The ENDF/B-VIII.0 evaluation of the $^{35}Cl(n,p)^{35}S$ reaction overestimates the cross section above 1.25 MeV and dramatically underestimates it below 1.25 MeV. Therefore, a full re-evaluation is recommended, along with new measurements at a wider range of energies and validation using detector systems like CLYC.

- Significant room for improvement:
  - Measurements at both Lujan(moderated Tungsten target) and WNR (unmoderated Tungsten target) to cover a wide range of energies with a consistent experimental setup.
  - NaCl or AgCl on Pt or Au backing foils
  - Additional angular coverage

- "Nonstatistical fluctuations in the $^{35}Cl(n,p)^{35}S$ reaction cross section at fast-neutron energies from 0.6 to 6 MeV" published in Phys. Rev. C. 102, 024623 (2020)

- To improve the fidelity of our MCNP and GEANT4 simulations, we have collaborated with evaluators at LANL and Dr. Kim of KAERI to develop improved evaluated data library inputs for (n,z) reactions guided by our LENZ reaction data on Fe, Ni, and brass target data. This work was recently published in Nuclear Instruments and Methods in Physics Research Section A, 964, 163699 (2020).