Neutron-Induced Reactions in a High-Density Inertial Confinement Plasma and Their Nuclear Astrophysics Nexus

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#### Heavy Isotope Abundances in Mike Thermonuclear Device\*

H. DIAMOND, P. R. FIELDS, C. S. STEVENS, M. H. STUDIER, S. M. FRIED, M. G. INGHRAM, D. C. HESS, G. L. PYLE<sup>†</sup>, J. F. MECH, AND W. M. MANNING Argonne National Laboratory, Lemont, Illinois

AND

A. GHIORSO, S. G. THOMPSON, G. H. HIGGINS, AND G. T. SEABORG Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California

AND

C. I. BROWNE, H. L. SMITH, AND R. W. SPENCE Los Alamos Scientific Laboratory, Los Alamos, New Mexico (Received May 2, 1960) The High-Density Plasma at National Ignition Facility

The closest analog to *explosive stellar conditions* in the laboratory and in particular the closest laboratory *neutron analog to astrophysical r-process* 



A. Zylstra et al. (2022) H. Abu-Shawareb et al (2024)

Ch. J. Cerjan et al. (2018)

#### N221204: ignition achieved 3.1 MJ fusion yield / 2.05 MJ 351nm laser light

#### experimental neutron tomographic reconstruction



Abu-Shawareb et al., PRL (2024)













Discovery Science proposal P-000523: "A noble-gas accelerator mass spectrometry platform at NIF for nuclear astrophysics" :

introduce seeds of Ar in DT fuel and observe (*n*,2*n*) and neutron capture reactions

Shot N221002 192 laser beams 1.2 MJ, 390 TW



Discovery Science proposal P-000523: "A noble-gas accelerator mass spectrometry platform at NIF for nuclear astrophysics" :

# introduce seeds of Ar in DT fuel and observe (*n*,2*n*) and neutron capture reactions



<sup>42</sup>**Ar** motivation: <sup>40</sup>Ar(2n,γ)<sup>42</sup>Ar, a "mini-r" process

Choice of Ar:

- three long-lived neutron induced products: <sup>39</sup>Ar(268 y), <sup>41</sup>Ar(110 min), <sup>42</sup>Ar(33y)
- Ar is a noble gas: can be reliably collected using a stable <sup>38</sup>Ar carrier
- <sup>39,42</sup>Ar detection at ultra-high sensitivity by atom counting

A hypothesis: could neutron capture occur on an excited/isomeric state?



See also: M. Winter <sup>41</sup>Ar MSc Thesis (2024), U. Jyvaskyla.

E (level) (keV)	XREF	J <sup>n</sup> (level)	T <sub>1/2</sub> (level)	<mark>Ε (γ)</mark> (keV)
0	ABCDEFG	7/2-	109.61  m  4 $8 \beta^- = 100$	
167.11 <i>9</i>	ABCDEFG	5/2-	315 ps 15	167.1 <i>1</i>
515.77 15	ABCD FG	3/2-	0.26 ns 8	348.7 <i>2</i> 516.0 <i>3</i>
1033.94 16	ABCD FG	3/2+	5 ps +28-3	517.9 <i>3</i> 866.7 <i>2</i>

<sup>41</sup>Ar low-lying levels



<sup>42</sup>Ar

#### Atom counting with noble gas accelerator mass spectrometry (NOGAMS) at ATLAS (Argonne): a brief



#### NIF shot N221002: 22 Oct 2022

#### <sup>40</sup>Ar(n,2n)<sup>39</sup>Ar measured



# NIF shot N221002: 22 Oct 2022 <sup>42</sup>Ar detected (Nov 2024)





Ca41	Ca42	Ca43	Ca44	Ca45	Ca46
7/2-	0+	7/2-	0+	7/2-	0+
EC	0.647	0.135	2.086	β-	0.004
K40	K41	K42	K43	K44	K45
4-	3/2+	2-	3/2+	2-	3/2+
EC,β- 6,0117	6.7302	β-	β-	β-	β-
Ar39	Ar40	It nl	Ar42	Ar43	Ar44
7/2-	0+	THPT	0+	(3/2,5/2)	0+
β-	99.600	β-		3-	β·
C138 37.24 m	C139 55.6 m	C140 1.35 m	Cl41 38.4 s	C142 6.8 5	Cl43 335
2.	3/2+	2-	(1/2,3/2)+		
β-	β-	β-	β-	β-	β-

Presently considered scenario: (within 160 ps)

D +T  $\rightarrow n + \alpha$ : E<sub>n</sub> ~ 14 MeV n + T -> T + n:  $E_T \sim 10 \text{ MeV}$ tertiary reaction in flight T +  ${}^{40}Ar \rightarrow p + {}^{42}Ar : {}^{40}Ar(t,p) {}^{42}Ar$  $\sigma$ (TALYS 2.03)) ~ 20 mb (not experimentally measured) Present scenario:

D+T -> n +  $\alpha$ : E<sub>n</sub> ~ 14 MeV tertiary reaction in flightn + T $-> t + n : E_T \sim 10 \text{ MeV}$ (within 160 ps) $T + {}^{40}\text{Ar} -> p + {}^{42}\text{Ar} : {}^{40}\text{Ar}(t,p) {}^{42}\text{Ar}$ 

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Ca41	Ca42	Ca43	Ca44	Ca45	Ca46
7/2-	0+	7/2-	0+	7/2-	0+
EC	0.647	0.135	2.086	β-	0.004
K40 1.277E+9 y	K41	K42 12.360 h	K43 22.3 h	K44 22.13 m	K45 17.3 m
4	3/2+	2-	3/2+	2-	3/2+
EC,B	6.7302	β-	β-	β-	β-
Ar39 269 y	Ar40	Ar41 109.34 m	Ar42 32.9 v	Ar43 5.37 m	Ar44 11.87 m
7/2-	0+	7/2-	0+	(3/2,5/2)	0+
β-	99.600	β-		3-	β-
Cl38 37.24 m 2-	Cl39 55.6 m 3/2+	Cl40 1.35 m 2-	Cl41 38.4 s (1/2,3/2)+	C142 6.8 s	CI43 3.3 s
β. *	β-	B-	β-	B-	β-

	New-HIDRA	expl.
T:H:D	50:00:50	
DT-n Yield	2.01E+15	1.49e15(NIF monitors)
38Ar	4.64E+04	
39Ar	1.20E+10	2.8 e10 (NOGAMS)
41Ar	7.73E+06	2.0 e7 (γ spect., NIF)
42Ar	8.45E+05	1.6 e6 (NOGAMS)
40K	3.88E+07	
41K	1.39E+07	
42K	5.96E+06	
39C1	1.71E+05	
40C1	2.71E+08	
375	1.31E+08	

# Summary

- Quantitative determination of neutron induced reaction yields on <sup>40</sup>Ar seeds in a stellar-like high-density plasma
- 2. The origin of the <sup>42</sup>Ar events observed is attributed *mainly* to a charged-particle reaction in flight, first time observed:

 $d(t,\alpha)n(t,n)t({}^{40}Ar,p){}^{42}Ar$ 

3. The implosion of a pure DD capsule is considered to search for a two-neutron capture reaction.

## Thank you for your attention.

Preliminary results: N221002  ${}^{42}\text{Ar}/{}^{38}\text{Ar}$ = (1.0 ± 0.3) × 10<sup>-14</sup> N( ${}^{42}\text{Ar}$ ) = 1.6 ± 0.5 × 10<sup>6</sup> atoms

- 11 counts of <sup>42</sup>Ar from TR5 sample during a 53.5-hour run: (1.0±0.3) ct/5 h
- ✤ 0 <sup>42</sup>Ar counts for <sup>atm</sup>Ar during a 25.9hour run: < 0.2 ct/5 h</p>



# ILL2: <sup>42</sup>Ar NOGAMS calibration sample (2023)

<sup>42</sup>Ar/Ar (NOGAMS) =  $(1.09 \pm 0.12) \times 10^{-11}$ <sup>42</sup>Ar/Ar (activity+dilution) =  $(1.16 \pm 0.05) \times 10^{-11}$ 



NOGAMS September Monthly Update

September 24, 2024

Best-to-date two-dimensional half-sphere HYDRA simulation for N221002 (DT-gas loaded Ar)

	N221002				
	Subscale Symcap	Experiment	Simulation		
	DT-n yield	1.49E+15	3.34E+15		
	N221002 Subscale Symcap DT-n yield Peak Burn (ns) DSR (%) $T_{ion}$ (keV) Burnwidth (ps) Ar in gas fill 39Ar 41Ar 40Cl 42Ar 1.6(5) 38Ar 37S	6.87	6.87		
	DSR (%)	0.52	0.67		
	T <sub>ion</sub> (keV)	4.31	4.02		
	Burnwidth (ps)	NA	162		
	Ar in gas fill	2.520E+15	2.522E+15		
	39Ar 2.8(	1)e+10 4.00E+10	3.28621E+10		
incompatible	41Ar	2.00E+07	7.80998E+06		
	<b>40Cl</b>	NA	4.70612E+08		
with neutron	42Ar 1.60	5)e+6 NA	0.0755 (i.e. 0)		
capture unles	S				
$\sigma \sim \text{few 100b}$	<b>38Ar</b>	NA	2.21030E+05		
	<b>37S</b>	NA	2.29266E+08		
	<b>39Cl</b>	NA	3.13837E+05		

	N221002	N221002		T/D		
	T/D=50/50	50/0/50	80/20	70/30	20/80	30/70
DT-n Yield	1.49E+15	2.01E+15	1.49E+15	1.69E+15	1.76E+15	2.43E+15
39Ar	2.8(1)E+10	1.20E+10	9.35E+09	1.04E+10	1.03E+10	1.46E+10
41Ar	2.00E+07	7.73E+06	6.23E+06	9.34E+05	6.82E+06	9.70E+06
42Ar	1.6(5)E+06	8.45E+05	1.01E+06	6.55E+06	3.31E+05	7.21E+05
				1		
			0	ur next shot		
			F	all 2025,		
			W	e'll be back.		

#### new-HYDRA simulations: T/D ratio study

#### D + T -> $\alpha$ + n(~14 MeV) 14 MeV neutron capture on <sup>40</sup>Ar





Discovery Science proposal P-000523: "A noble-gas accelerator mass spectrometry platform at NIF for nuclear astrophysics" :

# introduce seeds of Ar in DT fuel and observe (*n*,2*n*) and neutron capture reactions

Sc39	Sc40 182.3 ms	Sc41 596.3 ms	Sc42 681.3 ms	Sc43 3.891 h	Sc44 3.927 h	Sc45	Sc46 83.79 d	Sc47 3.3492 d
(//2-)	+- ΕCp,ΕCα,	EC	EC *	EC	EC *	* 100	4+ β-	β-
Ca38	Ca39	Ca40	Ca41	Ca42	Ca43	Ca44	Ca45	Ca46
0+	3/2+	0+	7/2-	0+	7/2-	0+	7/2-	0+
EC	EC	96.941	EC	0.647	0.135	2.086	β-	0.004
K37	K38	K39	K40	K41	K42	K43	K44	K45
3/2+	3+	3/2+	4-	3/2+	2-	3/2+	2-	3/2+
EC	EC	93.2581	EC,β-	6.7302	<u>β-</u>	R-	β-	β-
Ar36	Ar37	Ar38	Ar39	Ar40	Ar41	Ar42	Ar43	Ar44
0+	3/2+	0+	209 Y	0+	1 6,34 Ш 		(3/2,5/2)	0+
0.337	EC	0.063	3-	99.600	R-	B-	β-	β-
Cl35	Cl36	<b>Cl3</b> 7	Cl38	C139	Cl40	Cl41	Cl42	Cl43
3/2+	2+	3/2+	2-	3/2+	2-	(1/2,3/2)+	0.8 \$	5.5 8
75.77	EC,β-	24.23	β-	β-	β-	β-	β-	β-

#### **Collection** of reaction product atoms:

- inject a known volume of <sup>38</sup>Ar carrier in NIF chamber just after shot, pump chamber into a cryogenic trap system to separate Ar (Radiochemical Analysis of Gaseous Samples (RAGS) apparatus



#### The Radiochemical Analysis of Gaseous Samples (RAGS) apparatus



# N221002-001-999 RAGS FLTR RGA DT HDC Au, stable and rad carriers



## N221002 Ar sample



#### HYDRA simulation for a Ar-doped DT implosion at high power Ch. Cerjan, LLNL, private comm.

39Ar	2.706E+12
375	1.931E+10
41Ar	7.204E+08
39Cl	2.236E+08
40Cl	3.858E+10
38Ar	3.696E+09
365	2.080E+08
38Cl	2.456E+07
42Ar	1.550E+03

# HYDRA simulations: Ch. Cerjan (LLNL)

- The primary implosion experiment simulation tool used for NIF experimental design and analysis.
  - ALE (Adaptive Lagrange-Euler)
  - Finite element based (quadrilateral in 2D or hexahedral elements in 3D)
  - Massively parallel
  - PYTHON user scripts may be readily linked.
- Physics capabilities are extensive.
  - Consistent numerical treatment of the hydrodynamic equations, diffusive radiation transport, and diffusive electron conduction.
  - Substantial flexibility exists for different EOS and conductivity model choices.
  - Implicit Monte Carlo photon transport.
  - Particle Monte Carlo neutron, charged particle, and gamma-ray generation and transport.
  - In-line or post-processing radiochemistry available (KUDU).
- A static three-dimensional model exists that correlates implosion diagnostics and quantifies the stagnation properties.

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Best-to-date two-dimensional half-sphere HYDRA simulation for N221002 (DT-gas loaded Ar)

N221002	<b>NIF</b> diagnostics	
Subscale Symcap	Experiment	Simulation
DT-n yield	1.49E+15	3.34E+15
Peak Burn (ns)	6.87	6.87
DSR (%)	0.52	0.67
T <sub>ion</sub> (keV)	4.31	4.02
Burnwidth (ps)	NA	162
Ar in gas fill	2.520E+15	2.522E+15
39Ar		3.28621E+10
41Ar		7.80998E+06
<b>40Cl</b>		4.70612E+08
42Ar		0.0755 (i.e. 0)
38Ar		2.21030E+05
<b>37S</b>		2.29266E+08
<b>39Cl</b>		3.13837E+05

#### Atom counting with noble gas accelerator mass spectrometry (NOGAMS) at ATLAS (Argonne): a brief



# A test experiment: $^{39}\text{Ar}$ detection $^{38}\text{Ar}(n,\gamma)^{39}\text{Ar}$ with thermal neutrons from Soreq nuclear reactor





Auxilliary experiment: <sup>40</sup>Ar(n,2n)<sup>39</sup>Ar first cross section measurement (14 MeV) at TU Dresden/Helmholtz Zenter Dresden Rossendorf

 $T(D,n)^4$ He reaction at Rossendorf (HZDR) ~6.6(1)×10<sup>11</sup> 14 MeV ncm<sup>-2</sup> content : <sup>40</sup>Ar (5N) gas 4.5 hours

sphere: stainless steel volume: 4.18 cm<sup>3</sup> pressure: 20 bar

fast-neutron monitors: <sup>27</sup>Al, <sup>93</sup>Nb







#### Auxilliary experiment: <sup>40</sup>Ar(n,2n)<sup>39</sup>Ar first cross section measurement (14 MeV) at TU Dresden/Helmholtz Zenter Dresden Rossendorf



NOGAMS September Monthly Update

September 24, 2024

Best-to-date two-dimensional half-sphere HYDRA simulation for N221002 (DT-gas loaded Ar)

	N221002		
	N221002 Subscale Symcap DT-n yield Peak Burn (ns) DSR (%) $T_{ion}$ (keV) Burnwidth (ps) Ar in gas fill 39Ar 2.8 39Ar 2.8 39Ar 40Cl 42Ar 38Ar 37S	Experiment	Simulation
	DT-n yield	1.49E+15	3.34E+15
	Peak Burn (ns)	6.87	6.87
	DSR (%)	0.52	0.67
	T <sub>ion</sub> (keV)	4.31	4.02
	Burnwidth (ps)	NA	162
	Ar in gas fill	2.520E+15	2.522E+15
	39Ar 2.8(*	1)e+10 4.00E+10	3.28621E+10
measured by $\gamma$ spect,	41Ar (β <sup>-</sup> ,110 min)	2.00E+07	7.80998E+06
after shot, 41K line	<b>40Cl</b>		4.70612E+08
at 1294 keV	42Ar		0.0755 (i.e. 0)
	38Ar		2.21030E+05
	378		2.29266E+08
	<b>39Cl</b>		3.13837E+05



















after quantitative dilution with <sup>nat</sup>Ar:  $^{42}$ Ar/ $^{40}$ Ar (ILL2 calibration sample ): = (1.16 ± 0.05) × 10<sup>-11</sup>

ILL2 shipped to Argonne for NOGAMS

# ILL2 sample at Argonne: <sup>42</sup>Ar tuning







#### NIF sample N221002: 2023 experiment, same <sup>42</sup>Ar setting



# Nov '24 experiment: dedicated to <sup>42</sup>Ar search

#### **Timeline:**

-11/11-20: Ion source was run with UHP (Ultra-High Purity) Ar gas and <sup>40</sup>Ar<sup>8+</sup> monitored at ECR3 cup

-11/20-24: sample TR5B (NIF N221002), <sup>42</sup>Ar running, 53.5 h

-11/24-26: sample <sup>nat</sup>Ar (see below), <sup>42</sup>Ar running, 25.9 h

-11/26: ILL2 <sup>42</sup>Ar calibration sample, <sup>21</sup>Ne , <sup>42</sup>Ar running.



## Preliminary results summary

- <sup>42</sup>Ar unambiguously identified in sample N221002:

 $N(^{42}Ar) = 1.6 \pm 0.5 \times 10^{6}$  atoms

Extraneous presence of <sup>42</sup>Ar or crosstalk effects are ruled out.

-  $^{42}$ Ar calibration sample ILL2 OK: calibration (activity+dilution):  $^{42}$ Ar/ $^{40}$ Ar= (1.17 ± 0.05) × 10<sup>-11</sup>

NOGAMS:  ${}^{42}$ Ar/ ${}^{40}$ Ar= (1.09 ± 0.12) × 10<sup>-11</sup>

# <sup>42</sup>Ar: a "rare" nuclide

R. W. Stoenner, O. A. Schaeffer, S. Katcoff, Science (1965)

#### Half-Lives of Argon-37,

#### Argon-39, and Argon-42

Abstract. The half-lives of three argon isotopes have been carefully determined, with the following results:  $Ar^{37}$  $35.1 \pm 0.1$  days;  $Ar^{39}$ ,  $269 \pm 3$  years  $Ar^{42}$ ,  $32.9 \pm 1.1$  years. By combining the  $Ar^{42}$  value with earlier data, a cross section of  $0.5 \pm 0.1$  barn is calculated for the reaction, with thermal neutrons,  $Ar^{41}(n,\gamma)Ar^{42}$ .

Sc39 (7/2-)	Sc40 182.3 ms 4-	Sc41 596.3 ms 7/2-	Sc42 681.3 ms 0+	Sc43 3.891 h 7/2-	Sc44 3.927 h 2+	Sc45	Sc46 83.79 d 4+	Sc47 3.3492 d 7/2-
	ECp,ECα,	EC	EC *	EC	* EC	* 100	β-	β-
Ca38	Ca39	<b>Ca40</b>	Ca41	Ca42	<b>Ca43</b>	Ca44		<b>Ca46</b>
0+	3/2+	0+	7/2-	0+	7/2-	0+	7/2-	0+
EC	EC	96.941	EC	0.647	0.135	2.086	β-	0.004
K37	K38	K39	K40	K41	K42	K43	K44	K45
1.220 s 3/2+	7.050 m 3+	3/2+	1.277E+9 y 4-	3/2+	12.300 ft 2-	22.5 n 3/2+	22.15 m 2-	3/2+
EC	EC *	93.2581	EC,β- 0.0117	6.7302	β-	β-	β-	β-
Ar36	Ar37	Ar38	Ar39	Ar40	Ar41	Ar42	Ar43	Ar44
0+	3/2+	0+	209 y 7/2-	0+	7/2-	52.9 y 0+	(3/2,5/2)	0+
0.337	EC	0.063	β-	99.600	β-		3-	β-
Cl35	Cl36	<b>Cl37</b>	C138	Cl39	C140	Cl41	Cl42	C143
3/2+	3.01E+5 y 2+	3/2+	37.24 m 2-	3/2+	1.35 m 2-	(1/2,3/2)+	0.8 5	3.3 \$
75.77	EC,β−	24.23	β-	β-	β-	β-	β-	β-

<sup>42</sup>A is extremely rare in nature

<sup>42</sup>Ar/Ar in Earth atmosphere:  ${}^{42}$ Ar/Ar= 9.2  ${}^{+2.2}_{-4.6} \times 10^{-21}$  (Barabash et al., 2016)

Neutron-induced reactions in a high-density inertial confinement plasma at National Ignition Facility

- neutrons?
- high-density plasma?
- National Ignition Facility?
- proposal, experiment, results: a progress report