Subterranean production of neutrons, ^{39}Ar and ^{21}Ne: Rates and uncertainties

Ondřej Šrámek,[1] Lauren Stevens,[2] William F. McDonough,[2] Sujoy Mukhopadhyay,[3] R. J. Peterson[4]

1 Charles University in Prague; 2 University of Maryland; 3 University of California Davis; 4 University of Colorado Boulder `ondrej.sramek@gmail.com, www.ondrejsramek.net`

Motivation

Geochemistry — Underground production of neutrons, ^{39}Ar and ^{21}Ne depends on local **U, Th, K abundance**. Comparing predictions of underground noble gas' isotopic composition to newly available measurements can tell us about the source rock U, Th, K content.

Dark matter experiments — Argon detectors for WIMP searches require **low-radioactivity argon** (low ^{39}Ar), and there is ongoing effort driven by the experimental particle physics community to measure ^{39}Ar in **underground gas sources** at unprecedented sensitivity.

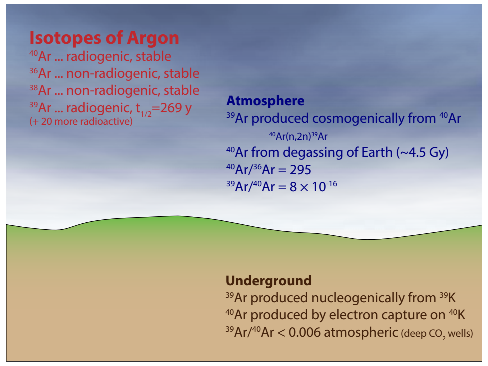
Hydrology — The radioactivity of ^{39}Ar (half life 269 years) produced in the atmosphere is used in **hydrological argon dating**. Knowledge of underground ^{39}Ar production rate is necessary for accurate age determination.

Curiosity — Existing calculations of nucleogenic ^{39}Ar production disagree and are unclear. Why?

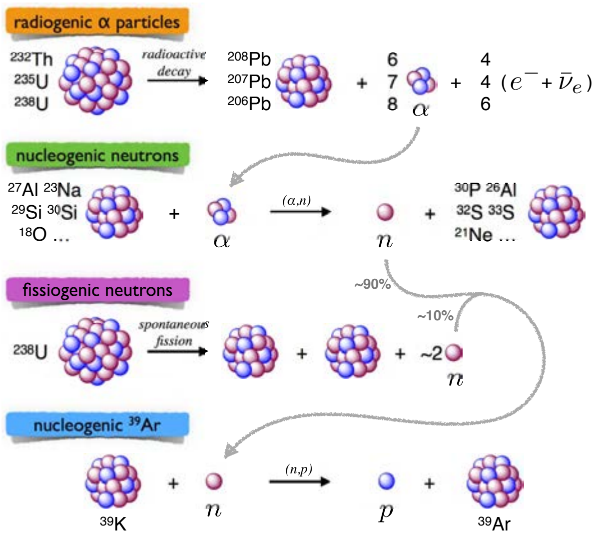
Goal

Calculate underground nucleogenic production rate of ^{39}Ar. Along the way, neutron and ^{21}Ne production rate are calculated.

1. Isotopes of argon



2. Nucleogenic ^{39}Ar production



3. Calculating ^{39}Ar nucleogenic production

Step 1: Alpha emission from decay of ^{232}Th and ^{238}U

- Decay chains contain several α-decays (Fig. 1).
- In each α-decay, α's can be emitted at several energy levels (Fig. 1).
- Emitted α particles slow down in material and eventually stop (Fig. 2).

Figure 1: *Decay networks and α emission spectra for ^{232}Th and ^{238}U. Spectra are normalized to number of α's emitted in decay chain (6 & 8).*

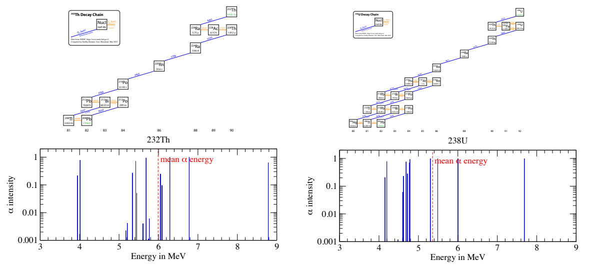
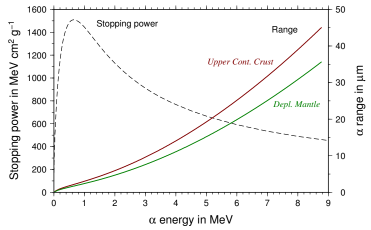


Figure 2: *Mass stopping power $-\rho^{-1}\mathrm{d}E/\mathrm{d}x$ for an α particle in rock (dashed line, left axis) and range of α particle (solid lines, right axis) as a function of α energy.*



Step 2: Neutron production by (α, n) reactions

- Before α stops, it can participate in an (α, n) reaction with a light target nuclide.
- Important target nuclides are ^{27}Al, ^{23}Na, ^{29}Si, ^{30}Si, ^{18}O, ^{26}Mg, ^{25}Mg.
- (α, n) cross sections calculated by `TALYS` version 1.6 (Fig. 3).
- *Neutron production function* in Fig. 4.
- Neutron energy spectra in Fig. 5.

Figure 3: *Energy dependent (α, n) cross sections for various target nuclides.*

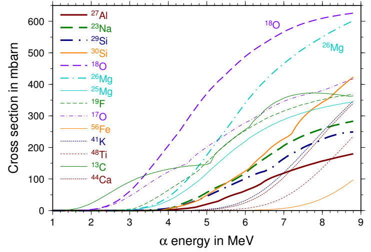


Figure 4: *Thick-target neutron production function: neutron yield per one α particle of a given initial energy.*

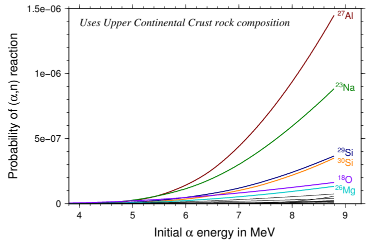
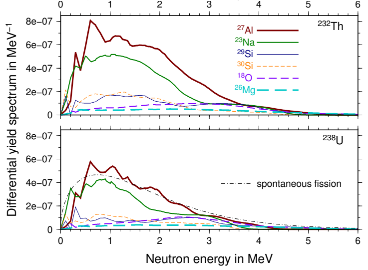


Figure 5: *Neutron spectra from α-induced reactions on various targets. Normalized to number of neutrons produced per 1 decay.*



Step 3: (n, p) and (n, α) reactions

Before the neutrons thermalize and get absorbed, they can participate in a number of reactions, including ^{39}K$(n,p)^{39}$Ar and ^{24}Mg$(n,\alpha)^{21}$Ne. The reaction yields were calculated with `MCNP6` code (`mcnp.lanl.gov`).

Data used: Decay data from "Chart of Nuclides" (NNDC @ BNL, `www.nndc.bnl.gov`). Stopping power from SRIM (`srim.org`). Neutron production cross section data calculated using `TALYS` code (`www.talys.eu`). Rock compositions from [1, 2, 3].

4. Calculated production rates

Table 1: *Calculated production rates of ^{4}He, neutrons, ^{21}Ne, ^{39}Ar, in number of atoms/neutrons per year per kg of rock.*

Composition	^{4}He	neutrons	^{21}Ne	^{39}Ar
Upper Cont. Crust	1.64×10^{10}	10680	753	28.7
Middle Cont. Crust	8.98×10^{9}	6114	416	13.9
Lower Cont. Crust	1.53×10^{9}	1129	70.2	0.749
Bulk Oceanic Crust	3.79×10^{8}	260	15.8	0.0235
Depleted Mantle	2.51×10^{7}	22.4	1.06	2.57×10^{-04}

Table 2: *Calculated production rates of ^{21}Ne by (α, n) and (n, α) and ^{21}Ne/^{4}He ratio.*

Composition	^{21}Ne prod. in atoms/kg-yr			% contrib.		
	(α, n)	(n, α)	Total	(α, n)	(n, α)	^{21}Ne/^{4}He
Upper Cont. Crust	753	0.159	753	99.98	0.02	4.59×10^{-8}
Middle Cont. Crust	415	0.165	416	99.96	0.04	4.63×10^{-8}
Lower Cont. Crust	70.1	0.104	70.2	99.85	0.15	4.58×10^{-8}
Bulk Oceanic Crust	15.8	0.0452	15.8	99.71	0.29	4.17×10^{-8}
Depleted Mantle	1.03	0.0366	1.06	96.55	3.45	4.23×10^{-8}

5. Uncertainty estimate of the calculation

Table 3: *Estimate of calculation uncertainty and its various contributions, assuming chemical composition (Upper Cont. Crust) is known precisely.*

	Error est. %
Decay data, α production	<1
Stopping power	3.5
Overall (α, n), neutron production	12
Overall (α, n), ^{39}Ar production	10
^{39}K(n, p) cross section	28
Neutron production calculation	13
^{21}Ne production calculation	17
^{39}Ar production calculation	30

6. Existing results for ^{39}Ar subsurface production

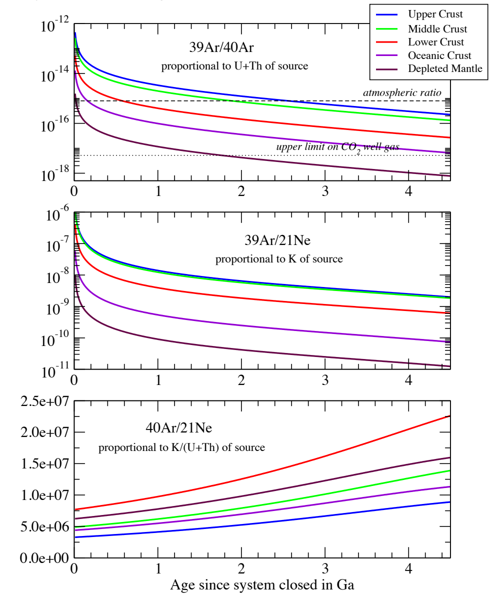
Table 4: *^{39}Ar production rates atoms per year per kg of rock from various studies. Recalculated to a common K, Th, U composition of Upper Cont. Crust.*

Reference	^{39}Ar prod. rate
Mei et al. 2010 [4]	11
Yokochi et al. 2012 [5]	55
Yokochi et al. 2013 [6]	170; 110
Yokochi et al. 2014 [7]	25; 25
This study	**29±9**

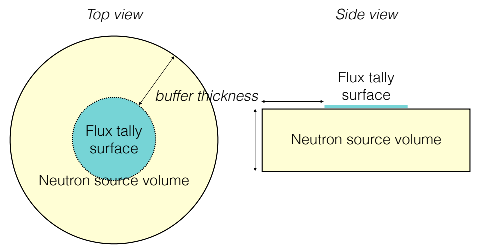
7. Constraints on K, Th, U abundances

- In a closed system, ^{21}Ne and ^{40}Ar (stable nuclides) accumulate while ^{39}Ar (half-life 269 years) reaches a steady state concentration.
- For a closed system of known age, measurements of various isotopic ratios can constrain the K, Th, U composition.

Figure 6: *Calculated noble gas ratio predictions for various rock compositions. Assumes system started degassed.*



8. Emission of nucleogenic + fissiogenic neutrons from a rock slab



Rock	Neutron flux
Granite (USGS G-2)	7.8×10^{-5} n/(cm^2 s)
Limestone (NIST SRM1c)	5.1×10^{-6} n/(cm^2 s)

The calculated nucleogenic + fissiogenic neutron fluxes from a rock into the air are 2–3 orders of magnitude below cosmogenically induced neutrons at sea level.

References

[1] Rudnick, R. L. and S. Gao, Composition of the continental crust, in R. L. Rudnick, editor, *The Crust*, volume 3 of *Treatise on Geochemistry*, chapter 3.01, pages 1–64, Elsevier Scientific Publishing Company, Oxford, 2003, doi:10.1016/B0-08-043751-6/03016-4, editors-in-chief H. D. Holland and K. K. Turekian.

[2] White, W. M. and E. M. Klein, Composition of the oceanic crust, in R. L. Rudnick, editor, *The Crust*, volume 4 of *Treatise on Geochemistry*, chapter 13, pages 457–496, Elsevier, Oxford, second edition, 2014, ISBN 978-0-08-098300-4, doi:10.1016/B978-0-08-095975-7.00315-6, editors-in-chief H. D. Holland and K. K. Turekian.

[3] Salters, V. J. M. and A. Stracke, Composition of the depleted mantle, *Geochem. Geophys. Geosyst.*, **5**(5):Q05B07, 2004, doi:10.1029/2003GC000597.

[4] Mei, D.-M., et al., Prediction of underground argon content for dark matter experiments, *Phys. Rev. C*, **81**(5):055802, 2010, doi:10.1103/PhysRevC.81.055802.

[5] Yokochi, R., N. C. Sturchio, and R. Purtschert, Determination of crustal fluid residence times using nucleogenic ^{39}Ar, *Geochim. Cosmochim. Acta*, **88**:19–26, 2012, doi:10.1016/j.gca.2012.04.034.

[6] Yokochi, R., et al., Noble gas radionuclides in Yellowstone geothermal gas emissions: A reconnaissance, *Chem. Geol.*, **339**:43–51, 2013, doi:10.1016/j.chemgeo.2012.09.037.

[7] Yokochi, R., et al., Corrigendum to "Noble gas radionuclides in Yellowstone geothermal gas emissions: a reconnaissance" [Chem. Geol. 339 (2013) 43-51], *Chem. Geol.*, **371**:128–129, 2014, doi:10.1016/j.chemgeo.2014.02.004.

Neutrino Geoscience 2015, 15–17 June, Institut de physique du globe de Paris, France