Cosmogenic activation in materials used in low background experiments

- Cosmogenic activation: origin and quantification
- Examples of activation studies:
 - Detector targets: Ar, Xe, Nal, Ge, others
 - Other materials: Cu, Pb, others
 - Underground activation



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Cosmogenic activation: origin



- **Primary cosmic rays:** ~90% p, 9% α, heavy nuclei, totally attenuated in the upper atmosphere
- Secondary cosmic rays on the Earth surface
 π[±]: p: e[±]: n: μ[±] observed with relative intensities
 1: 13: 340: 480: 1420





J.F. Ziegler, IBM J. Res. & Develop. 42 (1998) 1

Only μ survive >10 m.w.e: flux reduced by several orders of magnitude

Cosmogenic activation: origin

Production of long-lived radioactive isotopes in materials due to exposure to cosmic rays ("**cosmogenic activity**") can be an hazard for ultra-low background experiments

- On the *Earth's surface*, is dominated by **neutrons**; at *high altitude*, **proton** production is also important
- Muon activation deep underground can also be relevant
 - Limited knowledge of cosmogenic activation considered one of the "main uncertain nuclear physics aspects of relevance in the direct detection of dark matter".
 P. Gondolo, Nucl. Data Sheets120 (2014) 175
 - Relevant also in different contexts: Astrophysics, Geophysics, or Archaeology

Processes of nucleon-nuclei interaction

- Low energies: formation and decay of a long-lived **compound nucleus**
- GeV range: intranuclear cascade (INC) of nucleon-nucleon interactions followed by different deexcitation processes: spallation, fragmentation, break-up, fission, ...



Cosmogenic activation: mitigation

Avoid cosmic rays!

- Flights are forbidden
- Limit surface residency time during fabrication and transport of components
- Store, or even produce, materials underground Successful R&D for Ge crystal growth and detector fabrication D. Mei, arXiv:2409.03580
- Shielding against hadronic component of cosmic rays



This complicates the preparation of experiments \rightarrow it is desirable to have reliable **estimates of activation yields** to assess the real danger of cosmogenic activation

Cosmogenic activation: quantification

Recipe for estimates:

- 1. To know the **production rates** *R* of relevant isotopes in the targets, from
 - Scarce experimental data from irradiation / controlled exposure experiments
 - Calculations from production cross sections and cosmic ray spectrum

$$R = N_t \int \sigma(E) \phi(E) dE$$

- N_t = number of target nuclei ϕ = flux of cosmic rays σ = production cross section E = particle energy
- 2. To estimate the **induced activity** *A* knowing the **exposure history** to cosmic rays

$$A = R[1 - \exp(-\lambda t_{exp})] \exp(-\lambda t_{cool})$$

 t_{exp} = exposure time t_{cool} = cooling time underground

3. To compute the **background rate** generated by Monte Carlo simulation

Cosmogenic activation: flux of cosmic rays

At the Earth's surface nuclide production is dominated by **neutrons**

 \rightarrow A parametrization based on a set of measurements of cosmic neutrons on the ground across the US considered



Dependent on altitude (height of the atmosphere) and geomagnetic rigidity \rightarrow **correction factors** must be applied at different altitudes / latitudes

Cosmogenic activation: flux of cosmic rays

Other descriptions of the cosmic neutron spectrum are available



EXPACS (EXcel-based Program for calculating Atmospheric Cosmic-ray Spectrum): <u>https://phits.jaea.go.jp/expacs/</u>

to calculate terrestrial **cosmic ray fluxes** of **neutrons**, **protons**, light ions, **muons**, electrons, positrons, and photons nearly **anytime** and **anywhere** in the Earth's atmosphere.

CRY (Cosmic-ray Shower Library) generator

Cosmogenic activation: production cross sections

Select the best description of the excitation function $\sigma(E)$ by nucleons



- Experimental data: from beam experiments, typically few data for neutrons EXFOR (Experimental Nuclear Reaction Data) database

http://www-nds.iaea.org/exfor/exfor.htm



Cosmogenic activation: production cross sections

 Semiempirical formulae (Silberberg&Tsao equations): targets A ≥3, products A ≥ 6 and E>100 MeV

COSMO written in FORTRAN with three modes of calculation

- **Excitation curve** of a specified nuclide for a specified target
- Mass yield curve for given target and energy
- Activities produced for a target exposed to cosmic rays
- C. J. Martoff, P. D. Lewin, Computer Physics Comm. 72 (1992) 96

YIELDX FORTRAN routine to calculate the **production cross-section** of a nuclide in a particular target at a certain energy

- Including the latest updates of the Silberberg & Tsao equations (1998)

ACTIVIA C++ computer package to calculate

- Target-product cross sections
- Production and decay yields from cosmic ray activation

using semiempirical formulae but also experimental data tables if available

J. J. Back, Y. A. Ramachers, Nucl. Instrum. Meth. A 586 (2008) 286



Cosmogenic activation: production cross sections

- Monte Carlo simulation: standard packages (Geant4, FLUKA, ...) Specific codes for the interaction between nucleons and nuclei requiring the consideration of different reactions \rightarrow libraries

> GEM TALYS HMS-ALICE INUCL LAQGSM CEM ISABEL LAHET INCL+ABLA CASCADE MARS SHIELD BERTINI ...

TENDL (TALYS-based Evaluated Nuclear Data Library) https://tendl.web.psi.ch/tendl 2023/tendl2023.html

- Using the TALYS code
- For neutrons and protons up to 200 MeV

HEAD-2009 (High Energy Activation Data) https://doi.org/10.1016/j.nima.2010.08.110

- Only for Z≥12
- Using a selection of models and codes dictated by an extensive comparison with EXFOR data
- For protons from 150 MeV to 1 GeV
- **JENDL** (Japanese Evaluated Nuclear Data Library)
 - https://wwwndc.jaea.go.jp/jendl/jendl.html
 - Using GNASH code
 - For neutrons and protons up to 200 MeV, from 20 MeV to 3 GeV in High Energy File







Cosmogenic activation: quantification

The main sources of uncertainty in the evaluations come from difficulties on
precise evaluation of inclusive production cross-sections direct measurements needed to validate models
accurate description of cosmic ray spectra flux variation with latitude, longitude, altitude, and even time
precise exposure history tracking materials from fabrication to deployment

Snowmass2021 Cosmic Frontier White Paper: Calibrations and backgrounds for dark matter direct detection, arXiv:2203.07623

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Relevant cosmogenic products

 $^{39}\text{Ar:}\ \beta^{-}$ emitter with Q=565 keV, T_{1/2}=269 y mainly produced by $\ ^{40}Ar(n,2n)^{39}Ar$ Measured activity in

- **Atmospheric Ar:** ~1 Bq/kg (WARP, ArDM, DEAP)
- Underground Ar: (0.73±0.11) mBq/kg (DarkSide-50)

 $^{37}Ar:$ EC decay, E $_{\rm e,K\ shell}$ =2.8 keV, T $_{\rm 1/2}$ =35.02 d mainly produced by $^{40}Ar(n,4n)^{37}Ar$ Observed in early data of DarkSide-50

⁴²Ar: β⁻ emitter, Q=599 keV, T_{1/2}=32.9 y producing ⁴²K (β⁻ emitter, Q=3525 keV, T_{1/2}=12.36 h) → potential background for neutrinoless double beta decay

In Atm Ar: DBA: $92^{+22}_{-46} \mu$ Bq/kg, GERDA: 50-100 μ Bq/kg, DEAP (40.4±0.5.9) μ Bq/kg Production mechanisms: two-step neutron capture and 40 Ar (α ,2p) 42 Ar

Talks by Mario Schwarz and Niko Lay

³**H**: β^{-} emitter with Q=18.6 keV, **T**_{1/2}=12.3 y

• Purification systems for LAr should remove all non-noble radionuclides, as also assumed in **LXe**, but tritium proposed as a possible explanation for the XENON1T excess

Production rates R (sea level)

The Algorithm

First <u>measurement</u> for ³⁹Ar and ³⁷Ar in an irradiation experiment at Los Alamos (LANSCE) with a wide-band neutron beam that resembles the cosmic-ray neutron flux, quantifying products with a proportional counter at PNNL
 + calculations at sea level from alternate mechanisms

Talk	by Richard Saidanna			· · ·
Reaction	Estimated ³⁹ Ar production rate [atoms/(kg _{Ar} day)]	Fraction of total AAr (%)	Reaction	Estimated ³⁷ Ar production rate [atoms/(kg _{Ar} day)]
$\frac{{}^{40}\text{Ar}(n,2n)^{39}\text{Ar}+}{{}^{40}\text{Ar}(n,d)^{39}\text{Cl}}$	759 ± 128	72.3	$\frac{40}{40}$ Ar (n, 4n) ³⁷ Ar	51.0 ± 7.4
40 Ar (μ , n) ³⁹ Cl	172 ± 26	16.4	$\frac{Ar(\gamma, 3n)}{40}$ Ar (p, p3n) ³⁷ Ar	3.3 ± 0.7 1.3 ± 0.4
${}^{40} \text{Ar} (\gamma, n)^{39} \text{Ar} \\ {}^{40} \text{Ar} (\gamma, p)^{39} \text{Cl}$	$\begin{array}{c} 89\pm19\\ 23.8\pm8.7\end{array}$	8.5 2.3	36 Ar(n, γ) ³⁷ Ar	0.9 ± 0.3 (UAr) 36 ± 11 (AAr)
⁴⁰ Ar (p, 2p) ³⁹ Cl ⁴⁰ Ar (p, pn) ³⁹ Ar	< 0.1 3.6 ± 2.2	<0.01 0.3	38 Ar(n, 2n) ³⁷ Ar+ 38 Ar(ν , n) ³⁷ Ar+	<0.05 (UAr)
38 Ar(n, γ) 39 Ar	$\ll 0.1 (UAr)$ 1.1 $\pm 0.3 (AAr)$	- 0.1	$^{38}\text{Ar}(p, pn)^{37}\text{Ar}$	0.43 ± 0.05 (AAr)
Total	1048 ± 133	100		36.7 ± 7.5 (UAr) 92 ± 13 (AAr)

R. Saldanha et al, Phys. Rev. C 100 (2019) 024608

- Rates for ³⁷Ar, ³⁹Ar and ⁴²Ar from n, p and μ estimated by **GEANT4** simulation

C. Zhang, D.M. Mei, Astropart. Phys. 142 (2022) 102733

Production rates R (sea level): new calculations for DarkSide-20k

DarkSide-20k Collaboration, Astropart. Phys. 152 (2023) 102878; 2024 JINST 19 C02011



S. Cebrián, LRT2024, 2nd October 2024

Production rates R (sea level): new calculations for DarkSide-20k

DarkSide-20k Collaboration, Astropart. Phys. 152 (2023) 102878; 2024 JINST 19 C02011



R (atoms/kg/day)
115.1
177.2
221.6
168 ± 53
44.4
84.9
02.0

Activity A for DarkSide-20k from measured R for 37 Ar, 39 Ar and estimated R for 31 H assuming realistic exposure conditions at URANIA \rightarrow ARIA \rightarrow LNGS

Isotope	Activity (µBq/kg)
³⁹ Ar	20.7 ± 2.8
³⁷ Ar	103 ± 14
³ H (1)	76 ± 24
³ H (2)	2.97 ± 0.94

Tolerable residual level of 2.8% of quantified activity of ³⁹Ar in DarkSide-50

Each additional month at sea level would add 2.6 µBq/kg

Purification reduces activity of ³H by a factor 25

row (1) and (2): no / ideal purification

Large amounts of Xe being used in several huge DM and DBD experiments

	³ H	⁷ Be	¹²⁵ Sb	^{121<i>m</i>} Te	^{123m} Te	¹²⁷ Xe
Half-life [27,108]	12.312(25) y	53.22(6) d	2.75855(25) y	154 d	119.3(1) d	36.358(31) d
Measurement [184] Measurement [185]		32^{+21}_{-20}	51^{+22}_{-20}	<104	<53	$162^{+25}_{-23}\\132{\pm}26$
COSMO [184] ACTIVIA [184]		0.55 0.55	1.17 0.017	23.8 25.8	1.24 1.27	48.0 35.9
ACTIVIA [46] GEANT4 [46] TALYS [94]	35.6 31.6 16.0		$0.009 \\ 1.48 \\ 0.04$	54.5 21.2 11.7	2.67 18.5 12.1	89.9 233.3

Production rates in kg⁻¹d⁻¹

Controlled, long exposure to cosmic rays at LNGS and results from LUX L. Baudis et al, Eur. Phys. J. C 75 (2015) 485

³⁷Ar production by nuclear fragmentation of Xe quantified by LUX-ZEPLIN

- From Silberberg&Tsao cross sections + n, p spectra from Gordon and CRY
- Effect mitigated by Xe purification

 $R = 0.024 \text{ kg}^{-1} \text{ d}^{-1}$

J. Aalbers er al, Phys. Rev. D 105 (2022) 082004

Activation studies: Nal

 Cosmogenics found to make a very relevant contribution, according background models, in anual modulation DM experiments ANAIS-112 and COSINE-100

J. Amaré et al, Eur. Phys. J. C 79 (2019) 412; G. Adhikari et al, Eur. Phys, J. C 81 (2021) 837; G.H. Yu et al, arXiv:2408.09806



Detailed studies using data from experiments to quantify production of I, Te isotopes, ²²Na, ¹⁰⁹Cd, ¹¹³Sn in NaI(TI) crystals

J. Amaré et al, JCAP 02 (2015) 046, P. Villar et al, IJMPA 33 (2018) 1843006; E. Barbosa et al, Astropart. Phys.115 (2020) 102390



Activation studies: Nal

Production rates from known exposure and analysis of decaying signals



³H: additional background source required in the very low energy region



Measured rates
compared with
different calculations

Produc	:tion rates in kg	J ⁻¹ d ⁻¹	ANAIS		DM-Ice17
Isotope	$Half-life^{38,40}$	$Calculation^{33}$	$measurement^{33}$	$ACTIVIA^{34}$	$measurement^{34}$
^{126}I	12.93 d	297.0	283 ± 36	128	
^{125}I	$59.407 \ d$	242.3	220 ± 10	221	230
^{124}I	4.176 d	135.9			
$^{127m}\mathrm{Te}$	$106.1 {\rm d}$	7.1	10.2 ± 0.4	93	< 9
$^{125m}\mathrm{Te}$	$57.4 \mathrm{d}$	41.9	28.2 ± 1.3	74	27
$^{123m}\mathrm{Te}$	119.3 d	33.2	31.6 ± 1.1	52	21
$^{123}\mathrm{Te}$	$> 10^{13} \text{ y}$	10.2			
$^{121m}\mathrm{Te}$	$164 \mathrm{d}$	23.8	23.5 ± 0.8	93	25
$^{121}\mathrm{Te}$	19.16 d	8.4	9.9 ± 3.7	93	
113 Sn	$115.09 \ d$	9.9	6.8 ± 1.6	9.0	16
$^{109}\mathrm{Cd}$	$461.9 {\rm d}$	1.6	2.0 ± 0.6	4.8	
22 Na	2.6029 y	53.6	45.1 ± 1.9	66	

Activation studies: Nal



Results from **neutron beam irradiation** at Los

anha

Isotope	(atoms/kg/day)
³ H	80 ± 21
²² Na	47.8 ± 7.8
$^{109}Cd(+^{109}In + ^{109}Sn)$	5.7 ± 1.8
113 Sn(+ 113m Sn + 113 Sb + 113 Te)	7.1 ± 1.7
^{121m} Te	24.0 ± 3.9
^{123m} Te	30.5 ± 5.0
¹²⁵ I	271 ± 42
$^{125}Sb(+^{125}Sn)$	2.53 ± 0.55
^{125m} Te	125 ± 25
^{127m} Te	5.6 ± 4.4

Reasonable agreement between different estimates of **production rates** except for ^{125m}Te

- Widely used in detectors for DM, DBD, radioassay
- Cobalt isotopes are produced together with ⁶⁵Zn, ⁵⁴Mn and germanium isotopes (e.g. ⁶⁸Ge)

arXiv1802.09327 Counts [keV⁻¹.kg⁻¹.day⁻¹] - 68 Ge ^{₿8}Ge 10 68Ga 68Ga 657n 63NI 65Zn 57Co 55Fe 60Co 10-1 491 55Fe ⁴Mr 10-2 3H 49V 3H Total bkg 10-3 0^{-2} cn 10-4 10 16 18 12 10 14 Energy [keV] Simulation spectrum

Enriched Ge Production rates in kg⁻¹d⁻¹

	³ H	⁵⁴ Mn	⁵⁵ Fe	⁵⁷ Co	⁵⁸ Co	⁶⁰ Co	⁶⁵ Zn	⁶⁸ Ge
Half-life [27]	12.312(25)	312.19(3)	2.747(8)	271.81(4)	70.85(3)	5.2711(8)	244.01(9)	270.95(26)
units	У	d	У	d	d	У	d	d
Measurement [19]		2.3		1.6	1.2		11	
Measurement [103]		2.0 ± 1.0		0.7 ± 0.4		2.5 ± 1.2	8.9 ± 2.5	2.1 ± 0.4
Meas. (MAJORANA) [107]	140 ± 10	4.4 ± 4.1	2.1 ± 0.7				4.3 ± 3.6	3.3 ± 1.6
Monte Carlo [92]	140	1.4		1	1.8		6.4	0.94
Monte Carlo [93]				0.08	1.6	3.5	6.0	1.2
SHIELD [44]						3.3		5.8
TALYS [94]	24.0	0.87	3.4	6.7		1.6	20	7.2
MENDL+YIELDX [43]		3.7	1.6	1.7	4.6	5.1	20	12
TENDL+HEAD [28]	94 ± 34							
ACTIVIA [36]		2.2	1.6	2.9	5.5	2.4	10.4	7.6
ACTIVIA [45]	51.3	2.2	1.2	2.3	5.5	4.4	9.7	15.4
GEANT4 [45]	47.4	1.4	4.5	3.3	2.9	2.4	24.9	21.8
GEANT4+CRY [91]	22.8	0.96	2.9	2.8		1.9	18.0	20.0

Natural Ge

Production rates in kg⁻¹d⁻¹

	³ H	49 V	⁵⁴ Mn	⁵⁵ Fe	⁵⁷ Co	⁵⁸ Co	⁶⁰ Co	⁶⁵ Zn	⁶⁸ Ge
Half-life [27,108]	12.312(25)	330 d	312.19(3)	2.747(8)	271.81(4)	70.85(3)	5.2711(8)	244.01(9)	270.95(26)
units	у	d	у	d	d	у	d	d	
Measurement [92]			3.3 ± 0.8		2.9 ± 0.4	3.5 ± 0.9		38 ± 6	30 ± 7
Meas. (EDELWEISS) [100]	82 ± 21	2.8 ± 0.6		4.6 ± 0.7				106 ± 13	>71
Meas. (CDMSlite) [101]	74 ± 9			1.5 ± 0.7				17 ± 5	30 ± 18
Monte Carlo [92]	210		2.7		4.4	5.3		34.4	29.6
Monte Carlo [93]					0.5	4.4	4.8	30.0	26.5
Sigma [95]			9.1	8.4	10.2	16.1	6.6	79.0	58.4
SHIELD [44]							2.9		81.6
TALYS [94]	27.7		2.7	8.6	13.5		2.0	37.1	41.3
TALYS+INCL++-ABLA [101]	95			5.6				51	49
MENDL+YIELDX [43]			5.2	6.0	7.6	10.9	3.9	63	60
TENDL+HEAD[28]	75 ± 26								
ACTIVIA [36]			2.7	3.4	6.7	8.5	2.8	29.0	45.8
ACTIVIA [100]	46	1.9		3.5				38.7	23.1
ACTIVIA (MENDL-2P) [100]	43.5	1.9		4.0				65.8	45.0
ACTIVIA [45]	52.4		2.8	4.1	8.9	11.4	4.1	44.2	24.7
ACTIVIA [99]	30		3		6		3	20	10
GEANT4 [45]	47.4		2.0	7.9	7.4	5.7	2.9	75.9	182.8
GEANT4+CRY [91]	23.7	1.4	0.94	4.2	4.7		1.5	40.5	83.1
GEANT4+CRY [98]	21.6			2.9			0.9	27.7	63.6
CONUS [99]	50		5		7		4	60	66
CONUS experiment					9.0 ± 1.0			60 ± 10	200 ± 30

Nuclear Physics B - Proceedings Supplements Volume 28, Issue 1, July 1992, Pages 280-285

Theoretical and experimental investigation of cosmogenic radioisotope production in germanium

<u>F.T. Avignone IIII ⁽¹⁾, R.L. Brodzinski ⁽²⁾, J.I. Collar ⁽¹⁾, H.S. Miley ⁽²⁾, E. Garcia ⁽³⁾, A. Morales ⁽³⁾, J. Morales ⁽³⁾, R. Nuñez-Lagos ⁽³⁾, J.H. Reeves ⁽²⁾, C. Saenz ⁽³⁾, J.A. Villar ⁽³⁾</u>

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R. Breier, et al., Monte-Carlo calculation of production rates of cosmogenic radionuclides in a HPGe detector operating in the Modane underground laboratory, Nucl. Instrum. Meth. A 978 (2020) 164355, <u>http://dx.doi.org/10.1016/j.nima.2020.164355</u>.

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Q. Nie et al., Study of cosmogenic activation in ⁷⁶Ge enriched germanium detectors during fabrication and transportation above ground, 2024 JINST 19 P03002, <u>https://doi.org/10.1088/1748-0221/19/03/P03002</u>

Activation studies: other detector targets

Silicon: ³²Si, ³H

Controlled irradiation of silicon CCDs at Los Alamos

R. Saldanha et al, Phys. Rev. D 102 (2020) 102006

Talk by Richard Saldanha

Production rates in kg⁻¹d⁻¹

	³ H	⁷ Be	²² Na
Half-life [27]	12.312(25) y	53.22(6) d	2.6029(8) y
Measurement (neutrons) [232]	112 ± 24	8.1 ± 1.9	43.0±7.1
Measurement+Calculations (total) [232]	124 ± 24	9.4 ± 2.0	49.6 ± 7.3
TENDL+HEAD as [28]	120 ± 23		
TALYS+INCL++-ABLA [101]	124		k
GEANT4 [46]	27.3		
ACTIVIA [46]	108.7		



Activation studies: other detector targets

Tellurium:

p/n irradiations of TeO₂ at Los Alamos and CERN and study for ^{nat}Te

	⁶⁰ Co	^{110m}Ag	¹²⁴ Sb
Half-life [27]	5.2711(8) y	249.78(2) d	60.208(11) d
Measurement [136] ACTIVIA+TENDL [138]	<0.0053 0.070	0.42 0.206	15.7

A.F. Barghouty et al, Nucl. Instrum. Meth. B 295 (2013) 16 B. S. Wang et al, Phys. Rev. C 92 (2015) 024620

V. Lozza et al, Astropart. Phys. 61 (2015) 62

Production rates in kg⁻¹d⁻¹

Molibdenum:

Production rates in LMO of ⁸⁸Y, ⁸²Rb affecting double beta decay of ¹⁰⁰Mo Simulation based on **Geant4 + CRY** for n, p, μ and γ spectra

W. Chen, Eur. Phys. J. C 82 (2022) 549

CaWO₄:

Comparison of CRESST data and simulation based on Geant4 + ACTIVIA

H Kluck et al, 2021 J. Phys.: Conf. Ser. 2156 012227

Nuclide i	$^{14}\mathrm{C}$	$^{3}\mathrm{H}$	^{178}W	179 Ta	$^{175}\mathrm{Hf}$	^{181}W	$^{37}\mathrm{Ar}$	173 Lu	$^{172}\mathrm{Hf}$	^{171}Lu
$R_i / \mathrm{kg}^{-1} \mathrm{d}^{-1}$	84.06	34.82	25.91	24.59	16.67	13.74	12.17	11.25	11.12	9.18
TALYS		45.5				D. M	. Mei et al	, Astropart	. Phys. 31	(2009) 417

Activation studies: Tritium

Tritium can be a very relevant background in the detector medium of DM experiments due to its decay properties.



→ Specific study to quantify **production rates** induced in **targets** used in different **dark matter detectors: Ge, Si, Nal, Ar, Ne**

J. Amare et al, Astropart. Phys. 97 (2018) 96

$$R = N_t \int \sigma(E) \phi(E) dE$$

Activation studies: Tritium





Production rates in kg⁻¹d⁻¹

Target	Ref. 55	$\rm TENDL+\rm HEAD^{71}$	$TALYS^{59}$	GEANT4 ³⁷	GEANT4 ³⁶	ACTIVIA ³⁷	ACTIVIA ³⁶	ACTIVIA	Others
^{nat} Ge	178/210	75 ± 26	27.7	48.3	47.4	52.4	52.4	46/43.5 (Ref. 65)	82 ± 21 (Ref. 65)
									$76\pm 6~({\rm Ref.}~70)$
$^{\rm enr}{ m Ge}$	113/140	94 ± 34	24.0		47.4		51.3		140 ± 10 (Ref. 66)
Si		120 ± 23		27.3		108.7			125 (Ref. 52)
${\rm TeO}_2$			43.7						
NaI		83 ± 27	31.1	42.9		36.2	new meas	urements with	LANSCE bear
CsI			19.7				Si: 112 ± 2	4	
$CaWO_4$			45.5				R. Saldanha	et al, Phys. Rev	v. D 102 (2020)
Ar		146 ± 31	44.4	84.9		82.9	102006		
Ne		228 ± 16					Nal: 80 + 2	01	
Xe			16.0	31.6		35.6	R Saldanha	et al Phys Rev	D 107 (2023)
Quartz							022006	100 al, 1 11y3 100.	01 (2020)
$\mathrm{C}_{2}\mathrm{H}_{6}$				279.5			022000		

Cosmogenic activation in materials used in low background experiments

- Cosmogenic activation: origin and quantification
- Examples of activation studies:
 - Detector targets: Ar, Xe, Nal, Ge, others; ³H
 - Other materials: Cu, Pb, others
 - Underground activation



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Material largely used in experiments with many activation studies

	⁴⁶ Sc	^{48}V	⁵⁴ Mn	⁵⁶ Co	⁵⁷ Co	⁵⁸ Co	⁵⁹ Fe	⁶⁰ Co
Half-life[27,108] units	83.787(16) d	15.9735 d	312.19(3) d	77.236 d	271.81(4) d	70.85(3) d	44.494 d	5.2711(8) y
Measurement [202]	2.18 ± 0.74	4.5 ± 1.6	8.85 ± 0.86	9.5 ± 1.2	74 ± 17	67.9 ± 3.7	18.7 ± 4.9	86.4 ± 7.8
Measurement [184]	$2.33^{+0.95}_{-0.78}$	$3.4^{+1.6}_{-1.3}$	$13.3^{+3.0}_{-2.9}$	$9.3^{+1.2}_{-1.4}$	$44.8^{+8.6}_{-8.2}$	$68.9^{+5.4}_{-5.0}$	$4.1^{+1.4}_{-1.2}$	$29.4^{+7.1}_{-5.9}$
ACTIVIA (MENDL-2P) [36]	3.1	1.0	12.4	14.1	36.4	38.1	1.8	9.7
ACTIVIA [36,184]	3.1		14.3	8.7	32.5	56.6	4.2	26.3
COSMO [184]	1.5	3.1	13.5	7.0	30.2	54.6	4.3	25.7
ACTIVIA [46]	4.1		30.0	20.1	77.5	138.1	10.5	66.1
ACTIVIA [99]	3		16	9	34	60	2	29
GEANT4 [46]	1.2		12.3	10.3	67.2	57.3	8.8	64.6
TALYS [94]			16.2		56.2			46.4
MENDL+YIELDX [43]	2.7		27.7	20.0	74.1	123.0	4.9	55.4
CONUS [99]	3		14	10	50	76	5	92
Measurement (China)			18.6±2.0	9.9±1.3	48.3±5.5	51.8±2.5		39.7±5.7

Production rates in kg⁻¹d⁻¹

Measured rates from sensitive screening with Ge detectors after exposing large samples for long time in controlled conditions at **LNGS / Jinping** labs

S. Cebrián, et al., Cosmogenic activation in germanium and copper for rare event searches, Astropart. Phys. 33 (2010) 316–329, http://dx.doi.org/10.1016/j.astropartphys.2010.03.002.

L. Baudis, et al., Cosmogenic activation of xenon and copper, Eur. Phys. J. C 75 (2015) 485, http://dx.doi.org/10.1140/epjc/s10052-015-3711-3.

C. Zhang, et al., Cosmogenic activation of materials used in rare event search experiments, Astropart. Phys. 84 (2016) 62–69, http://dx.doi.org/10.1016/j. astropartphys.2016.08.008.

M. Laubenstein, G. Heusser, Cosmogenic radionuclides in metals as indicator for sea level exposure history, App. Rad. Isot. 67 (2009) 750–754, http://dx.doi. org/10.1016/j.apradiso.2009.01.029.

Z. She, et al., Study on cosmogenic activation in copper for rare event search experiments, Eur. Phys. J. C 81 (2021) 1041, http://dx.doi.org/10.1140/epjc/s10052-021-09827-w.

Stainless steel

Sample exposed for a long time at **LNGS** outside laboratory

Production	rates in	kg ⁻¹ d ⁻¹
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Isotope	⁷ Be	⁴⁶ Sc	48 V	⁵⁴ Mn	⁵⁶ Co	⁵⁸ Co
Half-life (d) [27,108]	53.22(6)	83.787(16)	15.9735	312.19(3)	77.236	70.85(3)
Measurement [202] GEANT4 [46] ACTIVIA [46]	$389 \pm 60 \\ 0.05 \\ 2.05$	$19.0 \pm 3.5 \\ 8.8 \\ 18$	34.6 ± 3.5	$233 \pm 26 \\ 230 \\ 191$	20.7 ± 3.5 16 131	51.8 ± 7.8 90 13

W. Maneschg et al. Nucl. Instrum. Meth. A 593 (2008) 448 M. Labustenstein, G. Heusser, ARI 67 (2009) 750

Production rates in kg⁻¹d⁻¹

Titanium		46 Sc	40 K
46Sc activity quantified by LUX	Half-life [27]	83.787(16) d	$1.2504(30) \times 10^9 \text{ y}$
	GEANT4 [46]	275.5	22.1
	ACTIVIA [46]	270.1	61.0

C. Zhang et al, Astropart. Phys. 84 (2016) 62

Activation studies: Pb, Ti, Al

Lead

• Sample exposed at Los Alamos to the neutron beam that resembles the cosmic-ray flux

 Activation previously unknown, found to be not relevant

Production rates in kg⁻¹d⁻¹

	¹⁹⁴ Hg	²⁰² Pb	²⁰⁷ Bi
Half-life (y) [27,108]	444	$5.25\ 10^4$	32.9
Measurement [205] TALYS [205]	$\begin{array}{c} 8.0 \pm 1.3 \\ 16 \end{array}$	$\begin{array}{c} 120\pm25\\77\end{array}$	< 0.17

V. E. Giuseppe et al, Astropart. Phys. 64 (2015) 34

Aluminium

Calculations based on different approaches, including measured production cross sections

B. Majorovits et al, Nucl. Instrum. Meth. A 647 (2011) 39 R. Breier et al, Nucl. Instrum. Meth. A 978 (2020) 164355

Production rates in kg⁻¹d⁻¹

	²² Na	²⁶ Al
Half-life (y) [27]	2.6029(8)	$7.17(24) \times 10^5$
Calculation for neutrons [209] Calculation for protons [209] ACTIVIA [99] CONUS [99]	153 24	389 47 160 530

Activation deep underground

Muons can produce by spallation radioisotopes inside the detector volume Effect of short-lived isotopes can be mitigated by time correlation with μ

¹¹**C** in **liquid scintillator:** three-fold coincidence between the crossing muon, the ejected neutron from ¹²C, and the ¹¹C decay (EC, β +, T_{1/2}= 20.4 m), allows a reduction of this background at the cost of a reduction of the life-time

Studies from irradiation experiments, data analysis of experiments like KamLAND and Borexino, and FLUKA simulations

T. Hagner et al, Astropart. Phys.14 (2000) 33

C. Galbiati et al, Phys. Rev. C 71 (2005) 055805

S. Abe et al, Phys. Rev. C 81 (2010) 025807

G. Bellini et al, J. Cosmol. Astropart. Phys. 08 (2013) 049

M. Agostini et al, Eur. Phys. J. C 81 (2021) 1075

^{77(m)}Ge in Ge detectors: decays of ⁷⁷Ge (β-, Q = 2.7 MeV, $T_{1/2}$ = 11.2 h) and metastable state ^{77m}Ge ($T_{1/2}$ = 53.7 s) affect ⁷⁶Ge DBD experiments Delayed coincidence cuts allow also to reduce this background.

C. Wiesinger et al, Eur. Phys. J. C 78 (2018) 597 M Neuberger et al, J. Phys.: Conf. Ser. 2156 (2022) 012216

Production of other metastable Ge isotopes quantified from Majorana data and simulations

I. J. Arnquist et al, Phys. Rev. C 105 (2022) 014617



Activation deep underground

Xe detectors: production rates of ³H, ¹³⁷Xe and other unstable Xe isotopes evaluated due to muon-induced neutron fluxes and spallation

- For four underground labs LNGS, SURF, LSM and SNOLAB for site selection of the DARWIN observatory
- Based on MUSIC-MUSUN + Geant4 simulation

M. Adrover et al, arXiv:2306.16340

Table 6 Muon-induced ¹³⁷Xe production rate at the different underground laboratories. The central value is the rate obtained with the Shielding physics list and the systematic error is calculated using the complementary simulations with the ShieldingLEND and QGSP_BICHP physics lists.

Site	Rate $(kg^{-1}yr^{-1})$
LNGS	$(8.22 \pm 0.27 \pm 1.00_{sys}) \cdot 10^{-4}$
SURF	$(1.42 \pm 0.12 \pm 0.21_{sys}) \cdot 10^{-4}$
LSM	$(1.65 \pm 0.11 \pm 0.30_{sys}) \cdot 10^{-4}$
SNOLAB	$(6.75 \pm 0.60 \pm 1.00_{sys}) \cdot 10^{-6}$

¹³⁷Xe (β ⁻ emitter, Q=4173 keV, T_{1/2}=3.82 m) from neutron capture analyzed for DBD

- From KamLAND-Zen: (1.42 ±0.73) 10⁻³ kg⁻¹ yr⁻¹
- Production rate at WIPP lab for EXO-200 from measured μ flux and Geant4 (FLUKA) simulations
 J. Albert et al, JCAP 04 (2016) 029
 439±17 (403±16) atoms per year
- Mitigation proposed by adding to Xe a small quantity of ³He to capture thermal n

L. Rogers et al, J. Phys. G 47 (2020) 075001

Activation deep underground

⁴²Ar in Ar: subsurface cosmogenic and radiogenic production carefully evaluated

Reactions	TALYS-	FLUKA	Major ⁴² Ar
	based pro-	residual-	yielding
	duction rate	nuclei-	reactions
	from selected	recording-	
	reactions	based)	
	[atoms/ton/y]	[atoms/ton/y]	
$n,p,\alpha,d,t-$	2.5×10^{-4}	4.2×10^{-4}	44 Ca $(n, 3$ He $)^{42}$ Ar
induced			
reactions			
Heavy-ion	_	8.3×10^{-4}	56 Fe(H*,X) 42 Ar
collisions			$^{44}Ca(H^*,X)^{42}Ar$
			$^{48}Ca(H^*,X)^{42}Ar$
Photon-	-	1.6×10^{-4}	44 Ca $(\gamma, X)^{42}$ Ar
induced			
reactions			
Pion-induced	—	1.6×10^{-4}	56 Fe(Π^-, X)
reactions			
Other	—	2.0×10^{-4}	${}^{44}Ca(\mu^-,2p){}^{42}Ar$
cosmic-ray			42 Cl β^- decay
muon-induced			
reactions			
Radiogenic	4.8×10^{-18}		$^{41}Ar(n,\gamma)^{42}Ar$
reactions			(, /)
All nooti	25×10^{-4}	1.9×10^{-3}	
All reactions	2.5 X 10	1.8 X 10	-
(sum)			

Isotope	Production rate in crust	Specific rad	ioactivi	ty in argon
	(atoms/ton (rock)/yr)	(decays/	ton (arg	gon)/yr)
³⁹ Ar	2.9×10^4 [15]	2.3	$\times 10^{7}$	7
⁴² Ar	1.8×10^{-3}		1.4	

S. Poudel et al, arXiv:2309.16169

- Standard continental crust, 3000 mwe
- Radiogenic contribution, based on TALYS cross sections, totally negligible
- Cosmogenic production in crust based on FLUKA simulation of μ's from MUSIC

 Activity in UAr gas evaluated from ³⁹Ar results, pointing to a suppression factor respect to AAr of at least 10⁷, much higher than for ³⁹Ar

Summary

Cosmogenic activation of materials can jeopardize the sensitivity of ultralow background experiments, being increasingly important as background requirements get more stringent

- production of long-lived isotopes at Earth's surface due to nucleons
- continuous generation of short-lived nuclides deep underground due to fast muons

Production rates and yields for several materials have been evaluated in the context of DBD, neutrino and DM experiments from direct **measurements** (with beams or from controlled, long exposure to cosmic rays) and from **calculations** based on different approaches

Mitigation is presently based on **limiting exposure** to cosmic rays but active suppression and removal of activation products can be considered:

- Underground crystal growth and detector fabrication (Ge, Nal)
- **Removal via post-processing** (cryogenic distillation for ³⁹Ar in Ar)



https://doi.org/10.3390/universe6100162

https://doi.org/10.1142/S0217751X17430060



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Article
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Cosmogenic Activation in Double Beta Decay Experiments

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MDPI

Abstract: Double beta decay is a very rare nuclear process and, therefore, experiments intended to detect it must be operated deep underground and in ultra-low background conditions. Long-lived radioisotopes produced by the previous exposure of materials to cosmic rays on the Earth's surface or even underground can become problematic for the required sensitivity. Here, the studies developed to quantify and reduce the activation yields in detectors and materials used in the set-up of these experiments will be reviewed, considering target materials like germanium, tellurium and xenon together with other ones commonly used like copper, lead, stainless steel or argon. Calculations following very different approaches and measurements from irradiation experiments using beams or directly cosmic rays will be considered for relevant radioisotopes. The effect of cosmogenic activation in present and future double beta decay projects based on different types of detectors will be analyzed too.

Keywords: neutrino; double beta decay; cosmic rays; activation; radioactive background