## 7. <sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl, and <sup>41</sup>Ca AMS Standards

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#### Introduction

Absolute AMS measurements require reliable standards for calibration. The author prepared <sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl, and <sup>41</sup>Ca AMS standards and distributed to many AMS laboratories. More than 60-90% of AMS measurements were normalized to these standards. These standards have been used as primary normalization standards at

University of Rochester
University of Pennsylvania
University of Tokyo
LLNL (Lawrence Livermore National Laboratory)

AMS standard is calibrated or defined by decay rate measurements or by isotopic ratio measurements of source material. If the standard was calibrated by decay rate, an uncertainty of isotopic ratio of the standard involves an uncertainty of the half-life. On the other hand, if we converted the measured isotopic ratio to activity, the result does not involve an uncertainty of the half-life. If the standard was calibrated by isotopic ratio measurements, number of atom in result is independent from an uncertainty of the half-life.

$$^{41}$$
Ca ( $t_{1/2} = 1.04 \times 10^5 \text{ yr}$ )

$$\frac{\frac{7/2^{-} 1.03 \times 10^{5} \text{ y}}{41 \text{ Ca}}}{\frac{41}{19} \text{ Ca}}$$

$$Q_{\text{EC}} 421.4$$

Half-life of <sup>41</sup> Ca		
$t_{1/2} (10^5 \text{ yr})$	Reference	
$1.03 \pm 0.04$	Mabuchi et al (1974)	
$(1.13 \pm 0.12)$	corrected	
$1.03 \pm 0.07$	Klein et al. (1991)	
$1.01 \pm 0.10$	Paul et al (1991)	
$1.04 \pm 0.05$	Mean value	

The preparation of <sup>41</sup>Ca AMS standards was published in Nishiizumi *et al* (2000). The highly enriched <sup>41</sup>CaCO<sub>3</sub> was obtained from ORNL (Oak Ridge National Laboratory). After purification of the CaCO<sub>3</sub>, the precise <sup>41</sup>Ca/Ca ratio in the original material was determined by surface ionization mass spectrometry.

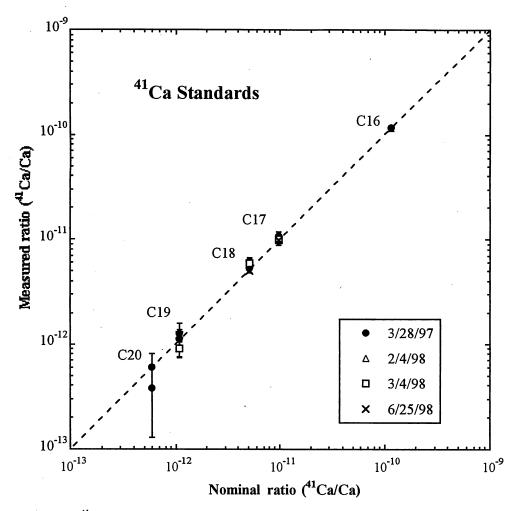
# Isotopic abundance measurements of enriched <sup>41</sup>Ca samples

Ca	UC Berkeley	Argonne*	CalTech*	ORNL
Isotope	(this work)			
40	97.79 ±0.02	97.77±0.06	97.75 ±0.09	97.76±0.06
41	1.2320±0.0002	$1.23\pm0.02$	1.237±0.001	1.33±0.04
42	$0.3620 \pm 0.0001$	0.34±0.01	$0.368 \pm 0.001$	0.34±0.03
43	Not measured	0.06±0.01	0.0376±0.0004	0.04±0.02
44	$0.5662 \pm 0.0001$	$0.52\pm0.01$	$0.555 \pm 0.001$	0.48±0.03
46	0.00030±0.00000	< 0.01	Not measured	<0.02
48	0.05360±0.00001	0.09±0.01	0.0491±0.0003	0.05±0.02

<sup>\*</sup> Paul et al.(1991)

(from Nishiizumi et al (2000))

The  $^{41}$ Ca was sequentially diluted with natural Ca to obtain  $^{41}$ Ca/Ca ratios of  $1.155 \times 10^{-10}$ ,  $9.755 \times 10^{-12}$ ,  $5.139 \times 10^{-12}$ ,  $1.095 \times 10^{-12}$ , and  $5.884 \times 10^{-13}$ . We estimated that an uncertainty of  $^{41}$ Ca concentration in stock solution is  $\pm 0.3\%$  (2 $\sigma$ ). Since the error of weighing and the concentration of Ca solution is estimated less than 0.1%, overall uncertainty of the  $^{41}$ Ca AMS standards that were diluted 5 times are less than 0.4%. The solutions having  $^{41}$ Ca/Ca ratios between  $10^{-10}$  and  $10^{-12}$  were measured by AMS at LLNL. Although the present blank level,  $5\times 10^{-14}$   $^{41}$ Ca/Ca, was higher than anticipated, excellent linearity was obtained from  $6\times 10^{-13}$  to  $1.2\times 10^{-10}$   $^{41}$ Ca/Ca.

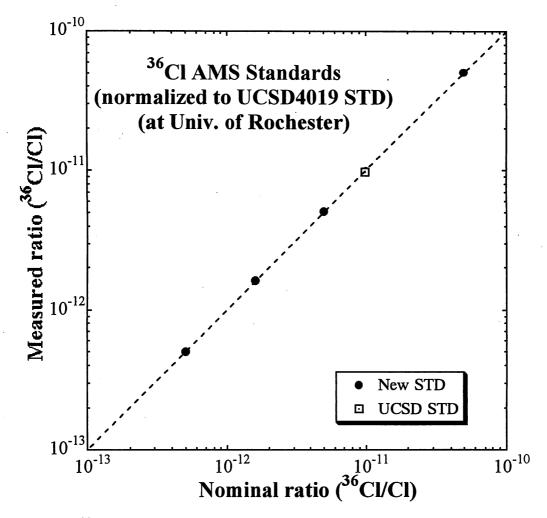


Results of <sup>41</sup>Ca standard measurements. All samples were normalized to C16, which has a nominal value of 1.155x10<sup>-10</sup> <sup>41</sup>Ca/Ca. (from Nishiizumi *et al* (2000))

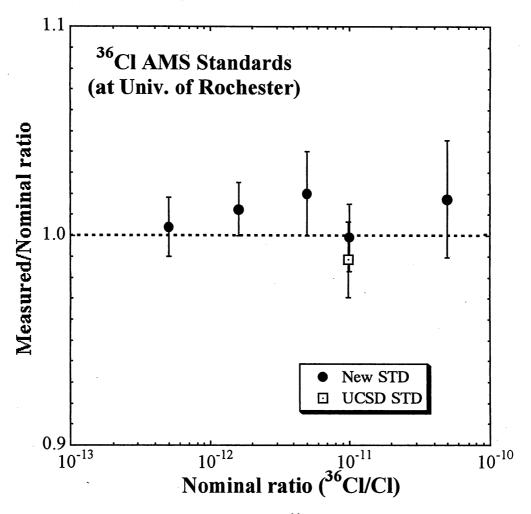
$$^{36}$$
Cl ( $t_{1/2} = 3.01 \times 10^5 \text{ yr}$ )

The preparation of <sup>36</sup>Cl AMS standards was published in Sharma *et al* (1990). Large-quantity dilutions of the NBS (National Bureau of Standards – present NIST) <sup>36</sup>Cl standard (SRM 4943) to <sup>36</sup>Cl/Cl ratios of 5.00x10<sup>-13</sup>, 1.600x10<sup>-12</sup>, 5.003x10<sup>-12</sup> and 1.000x10<sup>-11</sup> have been

prepared for AMS standards. Overall uncertainty in the activity was  $\pm 0.82\%$  ( $\pm 3\sigma$ ). Dilutions were made with an estimated uncertainty better than 1% and were then checked using the AMS facility at the University of Rochester. The results show excellent agreement between the gravimetric dilution factors and AMS measurements.

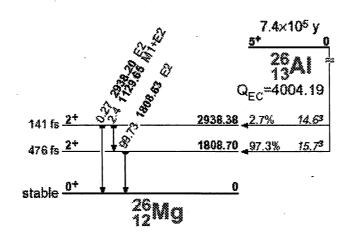


Results of <sup>36</sup>Cl standard measurements at University of Rochester. The x-axis indicates nominal value of each standard and y-axis indicates measured isotopic ratio of each standard that was normalized to old UCSD standard, which has a nominal value of 4.019x10<sup>-12</sup> <sup>36</sup>Cl/Cl.



Ratio of measured/nominal isotopic ratio of <sup>36</sup>Cl AMS standards prepared in 1983 and 1988. All standards are agreed well within uncertainties of AMS measurements.

$$^{26}$$
Al ( $t_{1/2} = 7.05 \times 10^5 \text{ yr}$ )



Half-life of <sup>26</sup>Al.

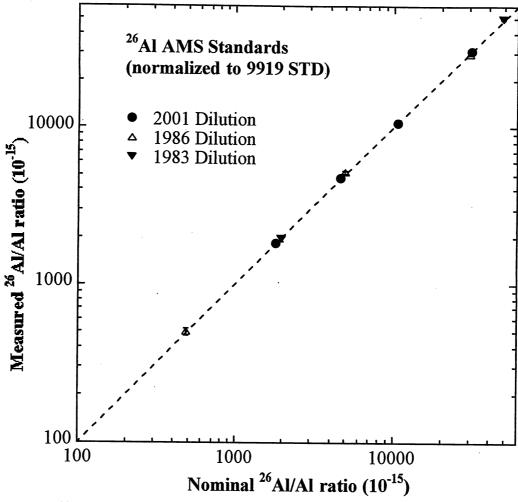
$t_{1/2} (10^5 \text{ yr})$	Method	Reference
$11 \pm 1$	γ_cross section	Rightmire et al. (1957)
$8 \pm 2$	γ_Mass spectrometry	Fisher et al. (1958)
7.38±0.29	4πγ Mass spectrometry	Rightmire et al. (1958)
7.16±0.32	Revised branching ratios	Samworth et al. (1972)
7.05±0.24	γ_Mass spectrometry	Norris et al. (1983)
6.99±0.56	Activity-AMS	Middleton et al. (1983)
7.05±0.56		, ,
$7.8 \pm 0.5$	γ_cross section	Thomas et al. (1984)

(from Nishiizumi (2003))

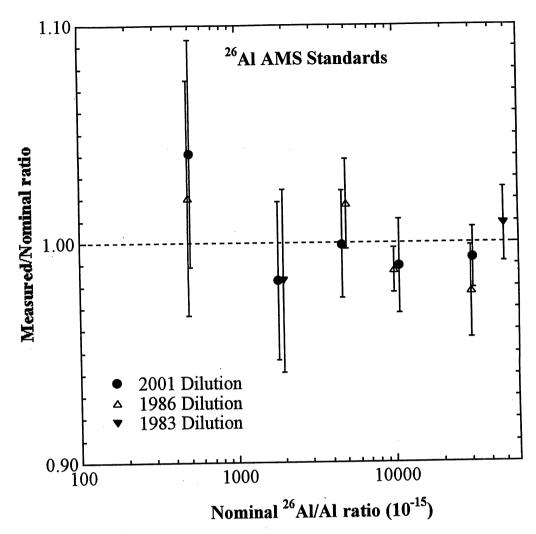
The preparation of <sup>26</sup>Al AMS standards is described in Nishiizumi (2003). Highly concentrated <sup>26</sup>Al was obtained from NBS (National Bureau of Standards). An uncertainty in the activity is 1.1% (3σ). The <sup>26</sup>Al, SRM 4229, was sequentially diluted with natural Al and <sup>26</sup>Al AMS standards were prepared. An overall uncertainty of the <sup>26</sup>Al AMS standards that were diluted 3 or 4 times is less than 1.2%. This is an uncertainty of specific activities. An uncertainty of absolute isotopic ratios (atom ratios) of AMS standards has to be included uncertainty of the half-life of <sup>26</sup>Al. These <sup>26</sup>Al AMS standards are widely used as primary standards at major AMS laboratories. A substantial amount of the <sup>26</sup>Al AMS standards, whose <sup>26</sup>Al/Al ratios are 7.444 x 10<sup>-11</sup>, 3.096 x 10<sup>-11</sup>, 1.065 x 10<sup>-11</sup>, 4.694 x 10<sup>-12</sup>, 1.818 x 10<sup>-12</sup>, and 4.99 x 10<sup>-13</sup> are now available for AMS community. Solutions having <sup>26</sup>Al/Al ratios between 10<sup>-12</sup> and 10<sup>-10</sup> were measured by several AMS laboratories; excellent linearity was obtained within the range of 5x10<sup>-13</sup> to 1x10<sup>-10</sup> <sup>26</sup>Al/Al.

An activity of the first dilution of our <sup>26</sup>Al standard solution was measured at the Max-Plank-Institute (MPI) Kernphysik against their <sup>26</sup>Al standard. The nominal value of our standard was 119.4±1.4 dpm based on NBS certificate and our dilution. The MPI-Kernphysik found an activity of 118.7±3.1 dpm, in excellent agreement with our nominal value (Heusser, pers. comm. 1986). Middleton *et al.* (1983) compared <sup>26</sup>Al standards from NBS and LMRI by AMS during the course of the <sup>26</sup>Al half-life measurements. They found that the concentration ratio of LMRI/NBS was 1.009; within errors, these standards are identical. The LMRI <sup>26</sup>Al standard is the primary <sup>26</sup>Al standard for AMS measurements for the Orsay group. Vogt *et al.* (Vogt *et al.*, 1994) compared our <sup>26</sup>Al standards with those prepared at Purdue University and found both standards were in excellent agreement, however, no detailed results were provided.

Wallner et al. (2000) compared <sup>26</sup>Al AMS standards from three different AMS laboratories using the VERA AMS facility. They found the isotopic ratio of the ETH-Zürich <sup>26</sup>Al standard differed by 13% relative to the others. The ETH-Zürich standard was also compared with our standard at LLNL. The nominal isotopic ratio of ETH-Zürich standard was 13 % higher than our standards as well. Although there are many different sources of <sup>26</sup>Al AMS standards, save for one, all <sup>26</sup>Al standards are undistinguishable from each other.



Results of <sup>26</sup>Al standard measurements at LLNL. The x-axis indicates nominal value of each standard and y-axis indicates measured isotopic ratio of each standard that was normalized to Al-86-4 standard, which has a nominal value of 9.919x10<sup>-12</sup> <sup>26</sup>Al/Al. (from Nishiizumi (2003))



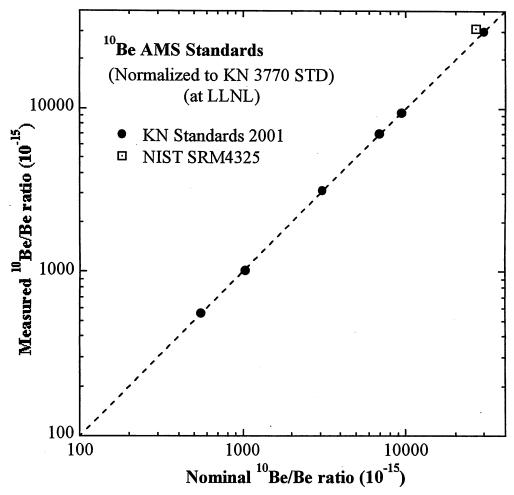
Ratio of measured/nominal isotopic ratio of <sup>26</sup>Al AMS standards prepared in 1983, 1986, and 2001. All standards are agreed well within uncertainties of AMS measurements. (from Nishiizumi (2003))

$$^{10}$$
Be  $(t_{1/2} = 1.5 \times 10^6 \text{ yr})$ 

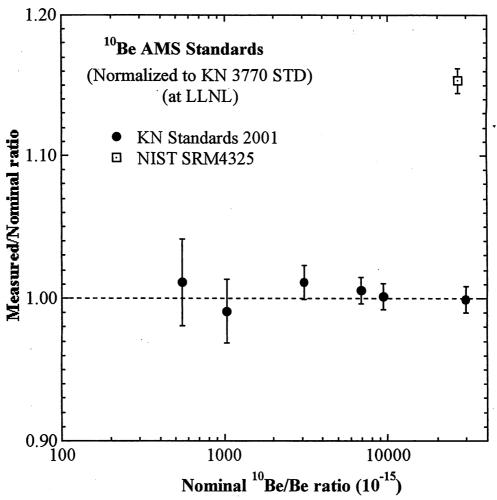
#### Half-life of <sup>10</sup>Be

$t_{1/2} (10^6 \text{ yr})$	Reference
1.7 ±0.4	McMillan (1947, 1972)
$1.6 \pm 0.2$	Emery et al (1972)
$1.5 \pm 0.3$	Yiou and Raisbeck (1972)
1.48±0.15	Makino et al (1975)
1.51±0.06	Hofmann et al (1987)
$1.34\pm0.07$	NIST SRM 4325 (1990)
1.48±0.06	Middleton et al (1993)

Highly concentrated <sup>10</sup>Be was obtained from ICN (ICN Chemical & Radioisotope Division). An uncertainty in the activity is 5%. The ICN <sup>10</sup>Be standard was sequentially diluted with natural Be and <sup>10</sup>Be AMS standards were prepared in 1983 and 2001. An uncertainty of absolute isotopic ratios (atom ratios) of AMS standards has to be included an uncertainty of the half-life of <sup>10</sup>Be. These <sup>10</sup>Be AMS standards are widely used as primary standards at major AMS laboratories. A substantial amount of the <sup>10</sup>Be AMS standards, whose <sup>10</sup>Be/Be ratios are 2.99 x 10<sup>-11</sup>, 9.42 x 10<sup>-12</sup>, 6.95 x 10<sup>-12</sup>, 3.11 x 10<sup>-12</sup>, 1.03 x 10<sup>-12</sup>, and 5.49 x 10<sup>-13</sup> are now available for AMS community. Solutions having <sup>10</sup>Be/Be ratios between 10<sup>-13</sup> and 10<sup>-11</sup> were measured by several AMS laboratories; excellent linearity was obtained within the range of 5x10<sup>-13</sup> to 3x10<sup>-11</sup> <sup>10</sup>Be/Be. However, the <sup>10</sup>Be/Be isotopic ratio of the ICN standards based on 1.5x10<sup>6</sup> yr half-life is 1.153±0.004 times higher than nominal value of NIST SRM 4325 standard, 2.68x10<sup>-11</sup>.



Results of <sup>10</sup>Be standard measurements at LLNL. The x-axis indicates nominal value of each standard and y-axis indicates measured isotopic ratio of each standard that was normalized to KN 3770 standard (prepared in 1983), which has a nominal value of 3.770x10<sup>-12</sup> <sup>10</sup>Be/Be.



Ratio of measured/nominal isotopic ratios of <sup>10</sup>Be AMS standards prepared in 2001 and NIST SRM 4325. All standards are agreed well within uncertainties of AMS measurements except NIST SRM 4325.

#### **Summary**

Large quantities of <sup>10</sup>Be, <sup>26</sup>Al, <sup>36</sup>Cl, and <sup>41</sup>Ca standards have been prepared for the AMS community. We recommend: (1) all AMS laboratories should note the source of the standards and half-life used to obtain their ratios; (2) the source of the standards and the half-life used in any AMS measurements should be clearly present in the publication; and (3) all sample calculations should use the same half-life as that of the primary normalization standard.

### Acknowledgement

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#### References

Emery J. F., Reynolds S. A., Wyatt E. I., and Gleason G. I. (1972) Half-lives of radionuclides - IV. Nucl. Sci. Eng. 48, 319-323.

- Fisher P. S., Hadley D. W., and Speers G. (1958) The positron decay of the ground state of aluminium-26. *Phi. Mag.* 3, 163-169.
- Hofmann H. J., Beer J., Bonani G., von Gunten H. R., Raman S., Suter M., Walker R. L., Wölfli W., and Zimmermann D. (1987) <sup>10</sup>Be: half-life and AMS-standards. *Nuclear Instruments & Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms* B29, 32-36.
- Klein J., Fink D., Middleton R., Nishiizumi K., and Arnold J. R. (1991) Determination of the half-life of <sup>41</sup>Ca from measurements of Antarctic meteorites. *Earth Planet. Sci. Lett.* 103, 79-83.
- Mabuchi H., Takahashi H., Nakamura Y., Notsu K., and Hamaguchi H. (1974) Half-life of <sup>41</sup>Ca. J. Inorg. Nucl. Chem. 36, 1687-1688.
- Makino T., Gensho R., and Honda M. (1975) Preparation of <sup>10</sup>Be and the measurement of isotopic ratio of beryllium. *Mass Spec.* 23, 33-39.
- McMillan E. M. (1947) Energy and half-life of the Be<sup>10</sup> radioactivity. Phys. Rev. 72, 591-593.
- McMillan E. M. (1972) Half-Life of <sup>10</sup>Be: A Correction. Phys. Rev. Lett., 2296.
- Middleton R., Klein J., Raisbeck G. M., and Yiou F. (1983) Accelerator mass spectrometry with <sup>26</sup>Al. *Nucl. Instrum. and Meth.* 218, 430-438.
- Middleton R., Brown L., Dezfouly-Arjomandy B., and Klein J. (1993) On <sup>10</sup>Be standards and the half-life of <sup>10</sup>Be. *Nucl. Instrum. and Meth.* B 82, 399-403.
- Nishiizumi K., Caffee M. W., and DePaolo D. J. (2000) Preparation of <sup>41</sup>Ca AMS standards. *Nucl. Instrum. and Meth.* 172, 399-403.
- Nishiizumi K. (2003) Preparation of <sup>26</sup>Al AMS Standards. Nucl. Instrum. and Meth., submitted.
- Norris T. L., Gancarz A. J., Rokop D. J., and Thomas K. W. (1983) Half-life of <sup>26</sup>Al. Proc. 14th Lunar Planet. Sci. Conf., J. Geophys. Res., B331-333.
- Paul M., Ahmad I., and Kutschera W. (1991) Half-life of <sup>41</sup>Ca. Zeitschrift für Physik A 340, 249-254.
- Rightmire R. A., Kohman T. P., and Allen A. J. (1957) Production of carrier-free aluminum-26 and sodium-22. J. Appl. Rad. Isotopes 2, 274-279.
- Rightmire V. R. A., Kohman T. P., and Hintenberger H. (1958) Über die Halbwertszeit des langlebigen <sup>26</sup>Al. Z. Naturforschg. 13, 846-853.
- Samworth E. A., Warburton E. K., and Engelbertink G. A. P. (1972) Beta Decay of the <sup>26</sup>Al Ground State. *Phys. Rev.* 5, 138-142.
- Sharma P., Kubik P. W., Fehn U., Gove H. E., Nishiizumi K., and Elmore D. (1990) Development of <sup>36</sup>Cl standards for AMS. *Nucl. Instrum. and Meth.* B52, 410-415.
- Thomas J. H., Rau R. L., Skelton R. T., and Kavanagh R. W. (1984) Half-life of <sup>26</sup>Al. *Phys. Rev.* C 30, 385-387.
- Vogt S., Wang M.-S., Li R., and Lipschutz M. (1994) Chemistry operations at Purdue's accelerator mass spectrometry facility. *Nucl. Instrum. and Meth.* B 92, 153-157.
- Wallner A., Ikebe Y., Kutchera W., Priller A., Steier P., Vonach H., and Wild E. (2000) Precision and accuracy of <sup>26</sup>Al measurements at VERA. *Nucl. Instrum. and Meth.* B 172, 382-387.
- Yiou F. and Raisbeck G. M. (1972) Half-Life of <sup>10</sup>Be. Phys. Rev. Lett. 29, 372-375.