

Direct determination of the half-life of ^{41}Ca . G. Jörg^{1,2}, Y. Amelin³, K. Kossert⁴ and C. L. v. Gostomski¹, ¹Radiochemie München (RCM), Technische Universität München, Walther-Meißner-Str. 3, 85748 Garching, Germany, ²Klinik und Poliklinik für Nuklearmedizin, Universitätsklinikum Würzburg, Oberdürrbacher Straße 6, 97080 Würzburg, Germany (joerg_g@klinik.uni-wuerzburg.de), ³Research School of Earth Sciences, Building 61, Mills Road, The Australian National University, Canberra ACT 0200 Australia, ⁴Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany.

Introduction: Radionuclides with half-lives between 10^5 and 10^8 years that were synthesized shortly before, or simultaneously with the formation of the Sun and our Solar System, are used to study the time-scale of accretion [1, 2] and the stellar environment of the Solar System's formation [3, 4]. The time resolution of these "extinct" nuclide chronometers depends on the half-life of the parent radionuclide: the shorter the half-life, the finer the bits of time that can be resolved. Since the time span from the formation of the central protostellar object to the pre-main sequence (classical T Tauri) stage of stellar evolution and formation of the protoplanetary disk is less than one million years [5, 6], extinct radionuclides with the shortest half-lives, suitable for resolving the fine temporal structure of this period, have a special value in the studies of the early Solar System.

^{41}Ca has the shortest half-life, about 10^5 years, among the radionuclides commonly used as "extinct nuclide" chronometers, but due to the progress of analytical techniques, the uncertainty of its half-life is increasingly becoming a limiting factor in chronological interpretations.

Numerous determinations of the half-life of ^{41}Ca were reported (Table 1). These data show that the half-life of ^{41}Ca is not reliably known: the values range from 7.7 to 19.0×10^4 years, and all values suffer from large uncertainties. Klein et al. [7] critically re-evaluated the published data and came up with substantially larger values for the half-life, and also used a novel alternative approach by linking content and activity to ^{36}Cl in Antarctic meteorites. Their methodology, however, also relies on critical assumptions about the process of $^{41}\text{Ca}/^{36}\text{Cl}$ production in the meteorites (i.e. saturation) and the sample history. Paul et al. [8] took one step closer to an analysis less parameter-dependent by determining the number of ^{41}Ca nuclei by mass spectrometry. However, they still carried the experimental drawbacks of X-ray measurements over into the result. As a result of methodological and experimental difficulties, all the reported values suffer from uncertainties as large as 10-30 %. Their low precision is an obstacle to the efficient use of this nuclide.

Here we report a new determination of the ^{41}Ca half-life with the set of techniques intended to achieve the best possible precision and accuracy.

Methods: Radiochemically pure ^{41}Ca was extracted from borosilicate glass from the absorber rods of a decommissioned pressurized water reactor. In order to maximize the $^{41}\text{Ca}/^{40}\text{Ca}$ ratio, calcium was separated without addition of inactive Ca carrier. The concentration of separated calcium and the $^{41}\text{Ca}/^{40}\text{Ca}$ ratio were measured by thermal ionization mass spectrometry with a ^{42}Ca - ^{48}Ca double spike, exponential normalization, and an independently determined absolute isotopic composition of Ca. The activity was measured by a liquid scintillation counting technique exploiting the triple-to-double coincidence ratio method, using the counting efficiencies computed following [9].

Results: The results of two independently processed and analysed samples of ^{41}Ca are presented in Table 2, together with a breakdown of uncertainty components. These two determinations yielded statistically indistinguishable values. Their average value of $(9.937 \pm 0.146) \times 10^4$ y is proposed as the new half-life value.

Discussion: The new half-life value overlaps the uncertainty interval of the previously accepted value of $(10.2 \pm 0.7) \times 10^4$ y [10], therefore it does not call for an immediate re-interpretation of the existing ^{41}Ca - ^{41}K data. Furthermore, the ^{41}Ca - ^{41}K method is still in the early stage of development and application, and the existing data are not particularly precise. With future refinements of the ^{41}Ca - ^{41}K techniques, the significance of a more precisely and accurately known ^{41}Ca half-life will increase.

References: [1] Kita N.T. et al. (2005) Chondrites and the Protoplanetary Disk. *ASP Conf. Ser.* 341, pp. 558–587. [2] Huss G.R. et al. (2009) *GCA* 73, 4922–4945. [3] Adams F.C. (2010) *Annual Rev. Astron. Astrophys.* 48, 47–85. [4] Dauphas N. and Chaussidon M. (2011) *Annual Rev. Earth Planet. Sci.* 39, 351–386. [5] André P. (2002) *Star Formation and the Physics of Young Stars*, pp. 1–38. Les Ulis, Fr.: EDP Sci. [6] Reipurth B. (2005) *Chondrites and the Protoplanetary Disk*. *ASP Conf. Ser.* 341, San Francisco, pp. 54–77. [7] Klein J. et al. (1991) *EPSL* 103, 79–83. [8] Paul M. et al. (1991) *Z. Phys. A* 340, 249–254 [9] Kossert K. and Grau Carles A. (2010) *Appl. Radiat. Isot.* 68, 1482–1488. [10] Cameron J. A. and Singh B. (2001) *Nucl. Data Sheets* 94, 429–603.

Table 1. The half-life values of ^{41}Ca reported in literature.

Reference	Half-life in 10^4 y	Relative standard uncertainty in %	Method
Sailor and Floyd (1951) ^a	---	---	First experiments with X-ray
Brown et al. (1953)	11.0(30)	27.3	n.-irr. of ^{40}Ca -enriched Ca ($n_{41\text{Ca}}$); X-ray ($A_{41\text{Ca}}$)
Drouin and Yaffe (1962)	7.7(11)	14.3	n.-irr. of Ca ($n_{41\text{Ca}}$); X-ray ($A_{41\text{Ca}}$)
Wahlin (1966) ^b	12.0	---	
Emery et al. (1972)	13.0(20)	15.4	
Mabuchi et al. (1974)	10.3(4)	3.9 ^g	n.-irr. of ^{40}Ca -enriched Ca ($n_{41\text{Ca}}$); X-ray ($A_{41\text{Ca}}$)
Browne and Firestone (1986)	10.3(4)	3.9	Decay data evaluation
Paul et al. (1991) ^{c, #, §}	10.1(10)	9.9	„standard MS techniques“ ($n_{41\text{Ca}}$); X-ray ($A_{41\text{Ca}}$)
Klein et al. (1991)	10.3(7)	6.8	Ratio $^{41}\text{Ca}/^{36}\text{Cl}$ in Antarctic meteorites
Klein et al. (1991) ^d	19.0(60)	32	Updated values for neutron capture cross section of ^{40}Ca and fluorescence yield of X-rays
Klein et al. (1991) ^e	16.0(25)	16	
Klein et al. (1991) ^f	11.4(5)	4.4 ^g	
Cameron and Singh (2001)	10.2(7)	6.9	Decay data evaluation
This work	9.937 (146)	1.47	TIMS and LSC

^(a) declare only “several months”

^(b) this work is quoted by Emery et al. (1972)

^(c) additional re-evaluations of earlier results in accordance with Klein et al. (1991)

^(d) re-evaluation of Brown et al. (1953)

^(e) re-evaluation of Drouin and Yaffe (1962)

^(f) re-evaluation of Mabuchi et al. (1974)

^(g) according to an evaluation by Kutschera et al. (1989), at least 30 % are more realistic

a complete report of the study tentatively described by Kutschera et al. (1989)

§ a recalculation by Kossert et al. (2009) yields 10.3×10^4 y

n.-irr.: neutron irradiation

$n_{41\text{Ca}}$: number of atoms of ^{41}Ca

$A_{41\text{Ca}}$: activity of ^{41}Ca

Table 2: Total uncertainty budget of the experimental data for calculation of the half-life of ^{41}Ca .

Component	Rel. uncertainty in %	Parameter	Sample No. 1	Sample No. 2
LSC	1.46	Total activity A ^{41}Ca in Bq	1529 ± 22	2076 ± 30
TIMS/Isotope dilution	0.08	Total content Ca in mg	1.360 ± 0.001	1.844 ± 0.001
		Specific Activity a ^{41}Ca in Bq/mg	1124 ± 16	1126 ± 16
TIMS	0.14	Isotopic concentration $n_{41\text{Ca}}$ in nmol/mg	8.452 ± 0.014	8.452 ± 0.014
Square root of the sum of quadratic components	1.47	Half-life in 10^4 y^d	9.946 ± 0.146	9.929 ± 0.146