Supplementary information to: New half-life measurement of ^{182}Hf: Improved chronometer for the early solar system

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CALCULATION OF THE ACTIVITY

The activity of ^{182}Hf, A_{182}, was calculated from the background-corrected peak area of the 270 keV line, N_{270}, the measuring time, Δt, the correction factor for the self-attenuation of the 270 keV γ-rays in the sample material, K_{270}, the absolute intensity of the 270 keV line, P_{270}, and the efficiency for 270 keV γ-rays, ϵ_{270}:

$$A_{182} = \frac{N_{270}}{\Delta t} \frac{K_{270}}{P_{270}\epsilon_{270}}. \tag{1}$$

The count rate of induced ^{181}Hf (r_{181} and r_{181_St}) was calculated from the background-corrected area of the full energy peak of the 482 keV γ-ray line, N_{482}. Corrections were applied for the decay between the end of the irradiation and the start of the measurement, ΔT, the decay during the γ-activity measurement, Δt, the self-attenuation of the 482 keV γ-rays in the sample material, K_{482}, and for the attenuation of the neutrons in the sample material during the irradiation, K_n:

$$r_{181} = N_{482}\lambda_{181} \frac{e^{\lambda_{181}\Delta T}}{1 - e^{-\lambda_{181}\Delta t}} K_{482} K_\mathrm{n}. \tag{2}$$

Correction factors for self-attenuation of γ-rays in the sample material, K_{270} and K_{482}, were based on the measured density and tables from energy dependent mass attenuation coefficients [1]. Due to the low concentration of Hf in the solutions prepared from material Helmer 2 (4 to 15 mg Hf in 3 ml) K_{270} was less than 0.1 % and thus negligible.

Correction factors for attenuation of the neutrons in the sample material during the irradiation, K_n, were calculated from the neutron capture cross section of the elements in the sample materials. Hf has a high cross section of 106 barn and thus was the only relevant component for the neutron attenuation in the high purity Hf standard materials. For Helmer 1 samples additional attenuation was included due to a significant contamination from boron stemming probably from the remaining original filter material (glass filter fibres). With a high neutron capture cross section of 760 barn boron critically influences the neutron attenuation corrections. The boron content was quantified to be (0.0128 ± 0.0003) g/g by comparing the induced ^{181}Hf activity of two neutron activations before and after chemical purification [2]. The uncertainty from this measurement was included in the final value for the half-life determined from material Helmer 1.

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[1] J. H. Hubbell and S. M. Seltzer, *http://physics.nist.gov/PhysRefData/XrayMassCoef/cover.html.*
[2] C. Vockenhuber *et al.*, Proceedings of 6th International Conference on Nuclear and Radiochemistry (2004).
[3] J. Wing *et al.*, Phys. Rev. **123**, 1354 (1961).

TABLES OF INDIVIDUAL MEASUREMENTS

TABLE I: Measured activities and calculated decay constants, λ_{182}, and half-lives, $t_{1/2}$, for individual measurements of Helmer 1 samples (uncertainties at 1σ). Series with identical measurement geometries are combined considering correlated uncertainties.

Sample	Measurement series[1]	A_{182} [2] (cts/s)	r_{181} [2] (cts/s)	r_{181_St}/n_{180_St} [2,3] (cts/s/10^{20} atoms)	λ_{182} [4] ($\times10^{-8}$ y^{-1})	$t_{1/2}$ ($\times10^6$ y)	Uncertainty statistical [4] ($\times10^6$ y)	Uncertainty total [5] ($\times10^6$ y)
3-8	3b	27.249 ± 0.218	0.05112 ± 0.00107	1.597 ± 0.029	8.563 ± 0.192	8.094	0.181	0.448
3-6	3	7.224 ± 0.242	0.01409 ± 0.00054	1.597 ± 0.029	8.239 ± 0.420	8.413	0.429	0.604
3-7	3	15.198 ± 0.285	0.02800 ± 0.00064	1.597 ± 0.029	8.719 ± 0.257	7.950	0.235	0.466
3-8	3	27.718 ± 0.197	0.05112 ± 0.00107	1.597 ± 0.029	8.711 ± 0.192	7.957	0.176	0.439
3-6	3b	7.337 ± 0.234	0.01465 ± 0.00059	1.597 ± 0.029	8.045 ± 0.411	8.616	0.441	0.620
3-6	4	7.352 ± 0.239	0.01405 ± 0.00245	1.617 ± 0.019	8.509 ± 1.507	8.146	1.442	1.496
3-7	4	15.246 ± 0.100	0.02752 ± 0.00305	1.617 ± 0.019	9.013 ± 1.002	7.690	0.855	0.933
3-8	4	27.274 ± 0.127	0.05253 ± 0.00108	1.617 ± 0.019	8.446 ± 0.177	8.206	0.172	0.435
5-7	5	17.302 ± 0.324	0.15934 ± 0.00365	7.659 ± 0.081	8.367 ± 0.247	8.284	0.245	0.470
5-8	5	42.042 ± 0.565	0.39537 ± 0.00253	7.659 ± 0.081	8.194 ± 0.122	8.460	0.126	0.428
					8.378 ± 0.414[6]	8.274	0.409[6]	0.447
3-6	6	7.027 ± 0.238	0.01405 ± 0.00245	1.617 ± 0.019	8.134 ± 1.442	8.522	1.457	1.502
3-7	6	13.808 ± 0.144	0.02752 ± 0.00305	1.617 ± 0.019	8.163 ± 0.909	8.491	0.912	0.950
3-8	6	24.851 ± 0.636	0.05253 ± 0.00108	1.617 ± 0.019	7.696 ± 0.252	9.006	0.285	0.343
5-7	6	16.322 ± 0.424	0.15934 ± 0.00365	7.659 ± 0.081	7.893 ± 0.273	8.782	0.293	0.328
5-8	6	38.514 ± 0.601	0.39537 ± 0.00253	7.659 ± 0.081	7.506 ± 0.127	9.235	0.150	0.202
					7.624 ± 0.164[6]	9.091	0.195[6]	0.280
3-8	7	25.292 ± 0.243	0.05253 ± 0.00108	1.617 ± 0.019	7.833 ± 0.177	8.850	0.285	0.258
5-8	7	38.938 ± 0.230	0.39537 ± 0.00253	7.659 ± 0.081	7.589 ± 0.066	9.134	0.113	0.134
					7.641 ± 0.118[6]	9.072	0.140[6]	0.244

[1] Measurement series 3, 4 and 5 were performed using the same measurement geometry (50% HPGe detector) and were γ-efficiency calibrated with a standard made from QCY46. Samples 3-6 and 3-8 were measured twice in series 3. Series 6 was measured using the same detector geometry, but calibrated with three preparations from a new calibration source QCY44. Series 7 was measured in a different geometry using a 30% HPGe detector with different detector electronics and γ-calibration with QCY44.
[2] Uncertainties comprise statistical uncertainties only.
[3] Combined induced ^{181}Hf count rate per ^{180}Hf of high purity Hf standards for respective irradiation series.
[4] Statistical uncertainties comprise statistical uncertainties of both A_{182} and r_{181}.
[5] Total uncertainties include in addition uncertainties from K_n, ϵ_{270} and P_{270}.
[6] Uncertainty includes also uncertainties from the respective ϵ_{270}.

TABLE II: Measured activities and calculated decay constants and half-lives for individual measurements of Helmer 2 samples (uncertainties at 1σ). Series with identical measurement geometries are combined considering correlated uncertainties.

Sample	Measurement series[1]	n_{182} [2] ($\times 10^{16}$ atoms)	A_{182} [3] (cts/s)	λ_{182} [3] ($\times 10^{-8}$ y^{-1})	$t_{1/2}$ ($\times 10^6$ y)	Uncertainty statistical [3] ($\times 10^6$ y)	Uncertainty total [4] ($\times 10^6$ y)
B1	2	1.223	23.838 ± 0.479	7.788 ± 0.157	8.900	0.179	0.211
B2	2	2.055	40.064 ± 0.485	7.788 ± 0.094	8.901	0.108	0.155
B3	2	2.861	55.655 ± 0.724	7.772 ± 0.101	8.918	0.116	0.161
B4	2	4.293	83.725 ± 0.541	7.790 ± 0.050	8.898	0.058	0.125
B4	2b	4.293	83.551 ± 0.728	7.774 ± 0.068	8.916	0.078	0.136
				7.784 ± 0.084[5]	8.905	0.097[5]	0.118
B1	10	1.223	23.849 ± 0.124	7.792 ± 0.041	8.896	0.046	0.107
B2	10	2.055	39.971 ± 0.152	7.770 ± 0.030	8.921	0.034	0.102
B3	10	2.861	56.061 ± 0.303	7.829 ± 0.042	8.854	0.048	0.107
B4	10	4.293	84.032 ± 0.454	7.819 ± 0.042	8.865	0.048	0.107
				7.795 ± 0.063[5]	8.892	0.071[5]	0.098

[1] Measurement series 2 was carried out with a 15% HPGe detector, series 10 with a 50% HPGe detector. Sample B4 was measured twice in series 2. For both series three standard solutions prepared from QCY44 were used for γ-efficiency calibration. Here all ^{182}Hf samples and γ-calibration standards were measured in identical solution volumes of 3 ml.
[2] Uncertainty from ID is 0.1%.
[3] Uncertainties comprise statistical uncertainties of A_{182} only.
[4] Total uncertainties include in addition uncertainties from ϵ_{270} and P_{270}.
[5] Uncertainty includes also uncertainties from the respective ϵ_{270}.

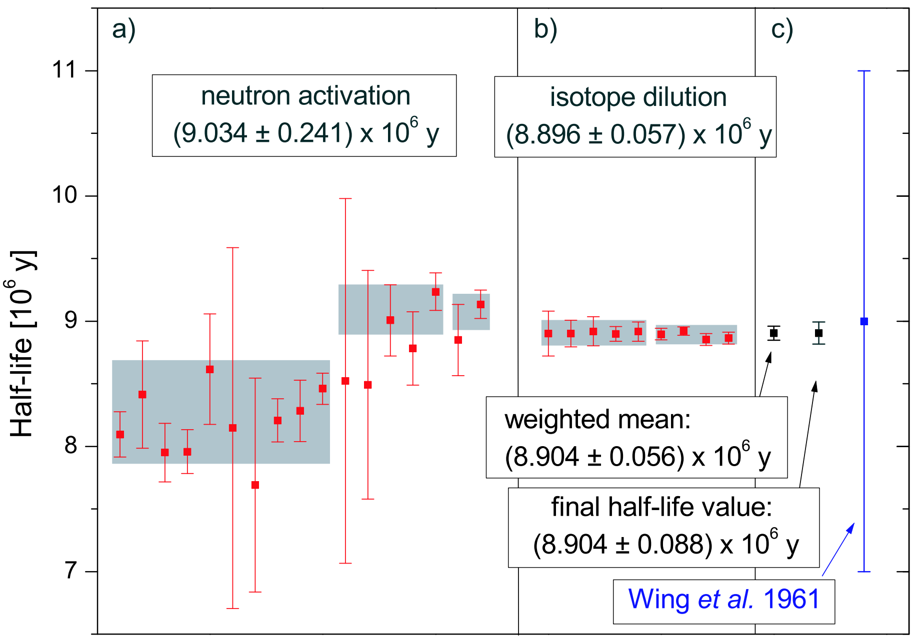

FIG. 1: Display of our various half-life measurements in chronological order, and their improvement with time. Individual data points in panels a) and b) show 1σ statistical uncertainties. The grey bars connecting individual measuring series (see Table I and II) mark their weighted mean $\pm 1\sigma$ ranges and include uncertainties from both statistics and efficiency calibration, the latter being the dominant error component. The boxed values labeled "neutron activation" and "isotope dilution" are the weighted means of the grey-bar areas representing Helmer 1 and Helmer 2 samples, respectively, which have been measured by independent methods. The value for the neutron activation measurements (Helmer 1) includes the uncertainty introduced by the correction for boron content. Panel c) shows the weighted mean of the two half-life values from a) and b) and our final half-life result, which includes the uncertainty of the γ-branch P_{270} in the ^{182}Hf-decay. The previous value published by Wing *et al.* [3] is also shown for comparison at the far right of the figure.