IUPAC-IUGS recommendation on the half-lives of ^{147}Sm and ^{146}Sm

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Abstract

The IUPAC-IUGS joint Task Group "Isotopes in Geosciences" recommends a value of (106.25 ± 0.38) Ga for the half-life of ^{147}Sm, and a corresponding decay constant $\lambda_{147} = (6.524 \pm 0.024) \times 10^{-12}$ a^{-1}, both with a coverage factor $k = 2$. For the extinct radionuclide ^{146}Sm two very different half-lives are used in the scientific community (*c*. 68 and 103 Ma), to such a degree that no consensus value can be endorsed at present by the Task Group. Pending dedicated re-investigations it is recommended that papers using the ^{146}Sm decay to quantify the cosmo/geological evolution of (extra)terrestrial samples perform a twin set of calculations using both proposed half-lives.

Introduction

The two isotopes of Sm most relevant for geochronological applications are ^{147}Sm and ^{146}Sm (Lugmair et al., 1975a, 1975b; Lugmair and Marti, 1977, 1978). Both are α emitters, which makes the discrimination of true disintegration events from noise straightforward. However, analytical challenges stem from the half-lives of these two isotopes: ^{147}Sm has a half-life, $t_{1/2}$, in excess of 100 Ga, resulting in a very low count rate, whereas ^{146}Sm has $t_{1/2} \leq 0.12$ Ga and is extinct in the Solar System, so that it must be synthesized before it can be counted. The α counting literature of the last 60 years includes 14 papers dealing with ^{147}Sm and four with ^{146}Sm. While all estimates agree on the order of magnitude, the individual half-life estimates differ by up to 30 %, despite one order of magnitude smaller published uncertainty estimates.

This situation requires a decision on how to distinguish acceptable measurements from incorrect ones. A first approach is to apply Gaussian statistics (e.g. Rajput and MacMahon, 1992) on the entire data-base from the literature, and then optimize the dispersion (quantified by the reduced chi square, χ^2_n, defined as the variance-weighted residuals normalized by the number of degrees of freedom, in the case of a weighted average the number of measurements minus one) by progressively removing measurements until a χ^2_n value acceptably near the expectation value of 1 is achieved. This statistical procedure identifies outliers, irrespective of the existence of experimental flaws. The χ^2_n is identical to the MSWD parameter (mean square of weighted deviates: McIntyre et al., 1966), widely used in geochronology. The requirement that χ^2_n be ≤ 1 is explained by the value of the *p* parameter, the "probability of fit". If $\chi^2_n > 1$, the dispersion of the measured points from the calculated ones (be it by linear regression or by averaging) is larger than the repeatability of the measurement; the data are then said to be overdispersed. This implies that the dispersion of values remains uncorrected for systematic errors, which include, possibly alongside other contributions, "incomplete knowledge of certain physical phenomena" (JCGM, 2008, §D.4). If $\chi^2_n > 1$, this also means that the likelihood, *p*, that the calculation accurately reflects the process that caused the distribution of the measured points becomes progressively smaller. The more typical examples

involve treating bimodal distributions as if they were unimodal, and linearly modeling a two-stage geological process as if it were single-stage. Since the likelihood that the function used for the calculation correctly predicts the observed dispersion is *p*, the chance that the calculation is incorrect is ~~by definition~~ 1-*p*; for a χ^2_n so high that $p = 0.05$ the chance of misidentifying the process controlling the distribution of the data points is 95 %.
An entirely different approach is to identify flawed experiments affected by systematic errors. To this end one aims to distinguish and evaluate the repeatability of measurements, typically via Type A evaluations (JCGM, 2012, entries 2.15 and 2.26), and the presence of systematic errors, typically via Type B evaluations "based on experience" (JCGM, 2012, entries 2.17 and 2.26). The IUPAC-IUGS Task Group has carried out Type B "expert" evaluations of the measurement protocols, and this is reflected in the present recommendation.

^{147}Sm

In the last 60 years a total of 14 counting experiments and one theoretical determination of the ^{147}Sm half-life have been published. What emerges is that one technique, liquid scintillation counting (LSC), gives four concordant measurement results (106.4 ± 0.8 Ga, with $\chi^2_n = 0.50$). Two other techniques appear at first sight to show a bimodal distribution, the ionization chamber and the silicon surface barrier. This bimodality suggests that one should look for a substantive explanation for this artifact rather than gloss over it by merely averaging all the measured values. In Appendix A, one exemplary dataset illustrates the paradoxical, and incorrect, conclusions of a pure Type A evaluation, in contrast to the Type B evaluation performed by the experimenters themselves.
The very authors of some experiments that have been shown to suffer from "substantive, documented" inaccuracy (Koepke et al, 2017a, §3.P1) had already set the corresponding results aside: (a) the Helsinki team (Valli et al., 1965) determined a half-life of 108 ± 2 Ga, superseding the earlier ionization chamber measurements by the same team (Karras and Nurmia, 1960; Graeffe and Nurmia, 1961); (b) Gupta and MacFarlane (1970) repeated, refined and replaced the earlier ionization chamber experiment by MacFarlane and Kohman (1961); (c) the silicon surface barrier measurement by Kinoshita et al. (2003) was later ignored by Kinoshita et al. (2012) who preferred using the 106 Ga half-life obtained in other experiments. These authors took great care to repeat their experiments eliminating factors that would lead to systematic errors after learning from past mistakes. It would be unfair (and statistically incorrect) to still include the superseded outliers into an unsupervised average by arbitrarily inflating the originally stated Type A uncertainty so as to transform a bimodal distribution with peaks around 116 Ga and 106 Ga (the 116 Ga peak being caused by incorrect experimental protocols) into a broader, unimodal Gaussian distribution, whose average and standard deviation offer no accurate information. The same argument applies to the use of Delrin plastic holders (see Appendix A), which were not a "random effect" to be propagated into an increased Dark Uncertainty, but pure and simply a flawed design, to which the argument made by Koepke et al. (2017b, p. 61, lines 7-8) must be applied. The remaining 10 measurements are listed in Table 1, grouped by experimental approaches. As the techniques are very different, any systematic error affecting one of them could not affect the other ones.
In addition to the counting experiments, Tavares and Terranova (2018) derived the half-life of ^{147}Sm from a first-principles calculation based on the quantum tunneling through a modelled potential barrier for the nucleus (Table 1). Any calculation relative to a nucleus with > 100 nucleons requires approximated models of multibody interactions; the accuracy of the model assumptions is not charted in all cases. What is certain is that a potential inaccuracy of the nuclear model would produce a bias completely unrelated to those that could have affected experimental studies. In the case of ^{147}Sm, the agreement with the experimental data is a very strong indication that no major inaccuracies affected the assumptions used for the calculation.
The fact that the results in Table 1 are mutually consistent is strong evidence that none of them is significantly biased. Indeed, an unintended benefit of the selection performed by the experimental groups themselves is that the statistical evaluation is that of a unimodal distribution (the reduced χ^2_n

= 0.28; Cochran's Q-test (Koepke et al., 2017a) yields a p-value of 0.99). Therefore, the conventional weighted average is appropriate, and yields a well-defined consensus value of 106.61 ± 0.99 Ga (k=2, for approximate 95 % coverage).

Table 1. Post-1958 direct determinations of the ^{147}Sm half-life. Only the 10 experimental papers not renounced by the original authors in later publications are considered. Uncertainties are taken from the original publication, with a coverage factor k = 1. LSC, liquid scintillation counter; IoC, ionization chamber; SSB, silicon surface barrier; Pro, proportional counter; Cr39, CR-39 detector. The four papers marked with an asterisk were those averaged by Lugmair and Marti (1978) to derive the half-life currently in use by the geochronological community, 106 ± 1.6 Ga. The original result by Martins et al. (1992) was corrected as explained by Begemann et al. (2001, p. 117). One theoretical calculation, marked as Calc, is also shown.

Method	reference	half-life (Ga)
LSC	Beard and Kelly (1958)	106 ± 4
LSC	Wright et al. (1961) *	105 ± 2
LSC	Donhoffer (1964) *	104 ± 3
LSC	Kossert et al. (2009)	107.0 ± 0.9
IoC	Valli et al. (1965) *	108 ± 2
IoC	Gupta and MacFarlane (1970) *	106 ± 2
IoC	Wilsenach et al. (2017)	107.4 ± 1.9
SSB	Su et al. (2010)	106.6 ± 0.8
Pro	Al Bataina and Janecke (1987)	105 ± 4
Cr39	Martins et al. (1992) **	106 ± 4
Calc	Tavares and Terranova (2018)	108 ± 3

Table 2. Determinations of the ^{147}Sm half-life by cosmo- and geochronological intercomparison. Comparisons based on meteoritic samples and terrestrial samples are labelled "met" and "terr", respectively. Only samples with acceptably low intra-sample dispersion, i.e. $\chi^2_n < 1$, are listed. The uncertainties, with a coverage factor k = 2, are either taken from the original publication, or recalculated from the original data and the U-Pb or Pb-Pb age of the same rock, propagating the uncertainty on the U half-lives (Villa et al., 2016).

Rock	reference for Sm-Nd age	reference U-Pb / Pb-Pb age	half-life (Ga)
terr	De Paolo and Wasserburg (1979)	Wall et al. (2016)	106.3 ± 0.9
met	Jacobsen and Wasserburg (1984)	Tissot et al. (2017)	105.8 ± 0.8
met	Lugmair and Galer (1992)	Amelin (2008)	106.1 ± 0.8
terr	Amelin and Semënov (1996)	Amelin et al. (1995)	106.4 ± 0.8

Cosmo- and geochronological intercomparisons are a half-life determination totally independent from alpha counting. Begemann et al. (2001) had concluded that geological intercomparisons are consistent with a (106.0 ± 1.6) Ga half-life (Lugmair and Marti, 1978) but not with the ca. 116 Ga half-life proposed in a part of the literature. The 106.0 Ga half-life is currently used by the geochronological community. In this evaluation we treat intercomparisons separately from counting experiments, as the metrological traceability only applies to the analytical part of the Sm-Nd and U-Pb (or Pb-Pb) age determination, but not to establishing the equivalence of the two age determinations. The equivalence is inherently a Type B evaluation (JCGM, 2012, § 2.29), as the intercomparison relies on the mineral phases being cogenetic, formed at the same time (or in a short time interval), and remaining closed to isotope exchange after formation; the evidence of such ideal "point-like" geological histories can only be established after their measurement. As some of the samples dated by Sm-Nd in the literature were also dated with other decay systems, the ideality of the point-like behavior can be better assessed. In the present evaluation we filtered the available cosmo- and geochronological intercomparisons by the requirement that the data define an overdetermined internal isochron having a statistically acceptable dispersion, i.e. $\chi^2_n \leq 1$, for all reported isotopic systems. This rigour is justified by the profound difference between "estimating the age of a sample" and "establishing a sample as an acceptable calibrator reference material for universal use". In the former case, most practitioners will accept some data overdispersion, even if $\chi^2_n > 1$ means that the likelihood that the result is accurate drops below 50 % (in regional geology studies it is sometimes tolerated that overdispersed isochrons having a near 0 % likelihood of accurately describing the distribution of data-points as due to a single-stage process be considered fit-for-purpose to the extent that they provide a first estimate of the approximate age of an otherwise undated sample). When the purpose of a measurement is dating a single sample, any inaccurate assumption only affects the accuracy of that sample. On the contrary, the selection of a reference material (JCGM, 2012, § 5.13) must be much more strict, as any systematic inaccuracy cascades on the accuracy on all measurements that rely on it (e.g. Potts, 2010). A geological intercomparison using natural rock samples as reference materials is only as good as the least homogeneous and the least pristine of the rocks in the comparison chain; therefore, the requirements for absence of any secondary modification must be much more strict than for general dating purposes.

Table 2 lists the intercomparisons from the literature that satisfy these *a priori* criteria. The weighted average of the four intercomparisons listed in Table 2 is $t_{1/2}$ = 106.22 ± 0.38 Ga, χ^2_n = 0.4. In summary, the weighted average of the determinations of the ^{147}Sm half-life listed in Tables 1-2 is (106.25 ± 0.38) Ga, with a coverage factor k = 2. The corresponding decay constant is λ_{147} = (6.524 ± 0.024) × 10^{-12} a^{-1}. The uncertainty on the half-life here recommended is about half of that of the most precise age of individual samples. In practical terms, the uncertainty is not completely negligible but is unlikely to become the dominant source of composite uncertainty with the present generation of analytical equipment. If and when the technical improvements will make it possible to reduce the measurement uncertainty consistently below that of the present recommendation, there will be scope for a renewed evaluation that will have to be based on the new, improved age intercomparisons - provided that χ^2_n is always < 1 for the selected overdetermined isochron calculations.

^{146}Sm

The number of publications reporting ^{146}Sm half-life measurements is quite small (Table 3). The most relevant problem is that the distribution of estimated half-life values is bimodal, whereby the two most discrepant half-lives estimates differ by a factor of 1.5. Two counting experiments give compatible results in the uncertainty interval (with a coverage factor k = 2) between 57 and 81 Ma, whereas two counting experiments agree in an uncertainty interval (with a coverage factor k = 2) between 93 and 112.2 Ma. This discrepancy needs to be addressed in detail.

Table 3. Determinations of the ^{146}Sm half-life, with a coverage factor k = 1. Four counting experiments are shown, with uncertainties taken from the original publications. The entry marked by an asterisk is the half-life calculation by Qian and Ren (2014), whose uncertainty estimate was not provided by its authors.

reference	half-life (Ma)
Nurmia et al. (1964)	74 ± 15
Friedman et al. (1966)	102.6 ± 4.8
Meissner et al. (1987)	103.1 ± 4.5
Kinoshita et al. (2012)	68 ± 7
Qian and Ren (2014)*	69

The quest for systematic artifacts is especially demanding. All papers state that they have estimated and taken into account all possible biases, but this obviously cannot be true for both contrasting modes of the bimodal distribution of half-life estimates. None of the early counting experiments actually provides a comprehensive description of all possible systematic artefacts. While limiting the descriptions of experimental details was commonplace in printed journals until the age of Electronic Supplements, it makes the evaluation of legacy papers nearly imposssible.

From the point of view of counting, one can note that the calculated half-life is inversely proportional to the number of detected events. Counting artifacts can include both false positives (detecting a signal caused by an interference and not corresponding to a genuine ^{146}Sm decay) and false negatives (not detecting a decay). Detecting a spurious 2.55 MeV signal is presumably unlikely; however, other possible artefacts can bias a counting experiment more severely than counting alone. Issues such as purity, stoichiometry and precise isotopic composition of the artificial ^{146}Sm source have not been adequately described by any of the four counting experiments listed in Table 3. Similarly to the recent "philosophical" shift that in U-Pb dating the non-isotopic information on petrogenesis is at least as important than mass spectrometer precision, the information on material purity and isotopic composition is at least as important as counting.

Friedman et al. (1966) followed two different experimental designs, so as to remove all artifacts that they considered: in one design, they determined the ^{145}Sm/^{146}Sm ratio, and in the other they "doped" ^{146}Sm with natural Sm. Meissner et al. (1987) were primarily interested in the systematics of alpha reduced widths and energies in a comparison of all nuclides with 84 neutrons; the half-life measurements of some of these nuclides were a by-product of the main goal of their experiments. Seeing as their estimate of the ^{146}Sm half-life was of the right order of magnitude and compared favorably with the two preceding literature results, they did not further discuss the possibility of artifacts that could have biased the half-life of just one among the nuclides that they were addressing, namely ^{146}Sm. Kinoshita et al. (2012) evaluated several possible causes for systematic error and concluded that none had affected their results significantly. They were first in addressing the possibility of isobaric interference of ^{146}Nd impurities during the mass spectrometric determination of the amount of ^{146}Sm. However, they did not document the stoichiometric purity of their Sm solution, nor did they determine its Sm isotopic composition by up-to-date analytical protocols.

Irradiated Sm targets were also used by Nurmia et al. (1964), who produced ^{146}Sm by ^{3}He irradiation of ^{147}Sm, and by Meissner et al. (1987), who produced ^{146}Sm via ^{146}Eu by ^{2}H irradiation of ^{147}Sm. Friedman et al. (1966) irradiated a Nd target to generate ^{146}Sm. In their experiment the ^{146}Sm half-life is independent from the Sm(dope)/Sm(sample) ratio. This implies that the supposed contaminant ^{146}Nd was not an impurity contained in the added Sm dopant, but probably was the result of ion exchange resins providing a less complete removal of the irradiation target than the 100.0000 % achieved by AMS (Kinoshita et al., 2012). The counting experiments should, in principle (Begemann et al. 2001), allow metrological traceability to a reference material having a known decay rate and a known mass fraction of the radioactive nuclide. Despite the foregoing explanation of possible discrepancies among the counting experiments, their very low number frustrates the attempt of a reliable evaluation of uncertainty.

The fifth entry in Table 3 is a theoretical calculation by Qian and Ren (2014) using a density-dependent cluster model. As these authors did not attempt to quantify the possible bias inherent in the model and did not provide an uncertainty estimate, their calculation cannot be compared with the counting experiments and cannot be evaluated in the present paper.

A number of studies proposed to evaluate the ^{146}Sm half-life by Early Solar System cosmo- and geochronological intercomparisons. A difficulty with such intercomparisons is that they all depend on very extensive assumptions on the ideality of single-stage evolution of the analyzed systems and on the uniform incorporation of one uniform ^{146}Sm/^{147}Sm ratio in the entire Solar System, none of which is amenable to a metrological tracing: there is neither a reference material for an extraterrestrial object with a single-stage point-like history, nor for its ^{146}Sm mass fraction, and therefore no propagation of uncertainty down the traceability chain. Mass spectrometric analyses *per se* are traceable to standard reference materials (as are the response functions of the gamma counters of the experiments in Table 3), but assumptions regarding the "point-like" history of early Solar System samples are not (just as the missing experimental details of the experiments in Table 3 are not). Traceability ensures that the measurement is accurate and that any other laboratory, when repeating the measurement under the same conditions, will reproduce the same number within the stated uncertainty. What traceability to an agreed reference material cannot guarantee is that the points in an isochron diagram pertain to a sample in petrologic equilibrium, nor can it guarantee that the isochron reflects the sample's single-stage formation age. A detailed discussion of results based on early Solar System samples is given in Appendix B.

There appears to be an unbridged gap between one part of the scientific community (e.g. Audi et al., 2017, p. 81), which only endorses the alpha-counting data (short half-life) measured by Kinoshita et al. (2012) and calculated by Qian and Ren (2014), versus another part of the community, which mostly accepts the long half-life measured by Meissner et al. (1987). At this time the inconsistencies evident in Table 3 cannot be pinpointed with certainty to a specific, documented systematic error. In the face of this dilemma, the Task Group must refrain from making a recommendation that satisfies both camps. If one seeks a consensus interval from the present data using the DerSimonian-Laird procedure as implemented in the NIST Consensus Builder (Koepke et al., 2017a, § 5.2), choosing the version of the uncertainty analysis that uses an adjustment (Knapp-Hartung) for the fact that the number of measurement results being combined is very small, and excluding the calculation by Qian and Ren (2014) because it is not qualified with an uncertainty evaluation, the resulting 95 % coverage interval ranges from 59 Ma to 119 Ma. This interval is too wide to be of much use; its width means that it is meaningless to try to blend such a heterogeneous set of measurement results into a single consensus value.

It is hoped that this review, by laying open all issues on undocumented systematic effects, will stimulate future work to improve the non-isotopic characterization (for the cosmochronological intercomparison) and non-counting characterizazion (for the counting experiments). A consensus on the half-life might only be reached if new, dedicated experiments will take into account the systematic errors identified over recent years. We recommend that until then all cosmo/geochronological reconstructions follow the interpretive approach by Sanborn et al. (2015, p. 92-93), who avoided firmly choosing between the 68 ± 7 Ma and the 103 ± 4 Ma half-life for ^{146}Sm and performed their calculations in double, comparing the results obtained using both conflicting assumptions.

Acknowledgements

The Task Group "Isotopes in Geosciences" is jointly funded by IUPAC (International Union of Pure and Applied Chemistry) and IUGS (International Union of Geological Sciences). Four anonymous reviews and one by R.W. Carlson were helpful. Associate Editor Yuri V. Amelin provided two rounds of extremely detailed, constructive criticism.

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Appendix A

Wilsenach et al (2017, their Table 3) describe two different types of holders on which the thin ^{147}Sm film is evaporated: metallic silicon and a plastic, Delrin. The five Delrin holder runs, taken alone, define a trimodal distribution. On these five samples, a Rajput-MacMahon-type statistical test confirms the inconsistency of the data set with a unimodal distribution, as $\chi^2_n = 11$. Removing successive points gave the best results for experiment SM004, with an average half-life of 117.1 ± 0.4 Ga and $\chi^2_n = 0.14$. The paradox is that this result is incorrect: pure Type A evaluations only take into account repeatability, irrespective of accuracy. However, the Type A approach was not pursued by Wilsenach et al. (2017), who instead correctly used a Type B argument. They realized that the isolating Delrin holders cause space charge artifacts, and performed later experiments with metallic Si holders. This modified design yielded a less precise but much more accurate half-life of 107.4 ± 1.9 Ga.

Appendix B

Most meteorites are contextless samples of parent planets whose geological history is unknown; even for meteorites from Moon, Mars and Vesta, the extraterrestrial bodies about whose evolution we have initial understanding, the data fall short of an uncontrovertible reconstruction of the local processes as would be required for using one of them as a metrologically acceptable standard calibrator.

The ages of several meteorite classes have been estimated by different short-lived chronometers, such as e.g. ^{26}Al-^{26}Mg, ^{53}Mn-^{53}Cr and ^{146}Sm-^{142}Nd, which give a reasonably consistent chronology of the first 5-10 Ma of Early Solar System events. The most useful samples for determining the ^{146}Sm half-life are however not the meteorites but rocks younger than 4.56 Ga by at least 0.2-0.3 Ga, because they provide a greater leverage on the rate of ^{146}Sm decay. Such samples are lunar highlands rocks and terrestrial Early Archean rocks. The post-accretion evolution of both Earth and Moon is very far from being known by unanimous consensus, let alone known with such accuracy to be useful as a metrological calibrator for the decay constant of ^{146}Sm. If samples with a relatively late formation age are chosen, their prehistory (including the presence of relicts and isotopic disequilibrium) and subsequent retrogression become the limiting factor for accuracy.

As discussed in the section on the ^{147}Sm half-life, we require that all internal isochrons respect $\chi^2_n \leq 1$ in order to be acceptable as half-life calibrators. This is a necessary requirement to ensure that the likelihood that an isochron represents the true single-stage evolution of the sample be greater than 50 %; if $\chi^2_n > 1$ this likelihood drops rapidly towards zero.

For meteorites there is evidence (the very title of the paper by Sanborn et al. (2015) explicitly alludes to it) that the parent planets of the meteorites in our collections had a prolonged history of primary and secondary events. These authors propose a secondary Nd isotopic disturbance (Sanborn et al., 2015, § 4.1) of the angrite parent planet around 4.51 Ga (Sanborn et al., 2015, p. 91), at a time the shorter-lived systems had long become extinct. It is therefore not obvious that the very short-lived systems would record the proposed 4.51 Ga disturbance. Moreover, since neither the geochemical mobility of parent elements Al, Hf and Mn is a proxy for that of parent element Sm, nor that of daughter elements Mg, W and Cr faithfully mirrors that of Nd, it is not rigorous to use the very short-lived radioactive Al-Mg, Hf-W and Mn-Cr systems to deduce the pristineness of the Sm-Nd pair. The issue is complicated even more by the suggestion by Tissot et al. (2017, p. 610-615), who list the possibilities of subtle (≤ 0.1 %) analytical artifacts in the U-Pb, Al-Mg and Mn-Cr chronometers in meteorites, implying that contemporaneity of samples and isotopic equilibrium in the early Solar System may be probable but are not certain. Indeed, all ^{146}Sm-^{142}Nd isochrons on the angrites analyzed by Sanborn et al. (2015, p. 84-88) were obtained after eliminating olivine and/or plagioclase to achieve χ^2_n values < 20 (but still mostly > 1).

Younger lunar samples were dated by the Carnegie-Livermore collaboration (Carlson et al., 2014; Marks et al., 2014, and abstracts quoted therein). Their primary interest was estimating the age of the magma ocean on the Moon (Carlson et al., 2014); the ages obtained in their survey are discussed by Carlson et al. (2014, p. 11) in the following verbatim quote:

> "*As with many studies of lunar highland samples, this study finds discordant ages for different isotopic systems when applied to exactly the same mineral and whole rock separates. Choosing which age best represents the crystallization age of the rock depends on applying subjective criteria. In reality, none of the ages need to reflect accurate crystallization ages as the lack of concordance from the different systems is a clear indication of disturbance of the radioisotope systems*."

This supports an open-system behavior (like that documented by Sanborn et al., 2015) and raises questions if the isochrons in the abstracts cited by Marks et al. (2014, Fig. 4) reflect a single-stage evolution of ^{142}Nd over time, i.e. only mirror the half-life of ^{146}Sm. The observation that angritic achondrite D'Orbigny exhibits

> *"surprisingly disturbed ^{147}Sm-^{143}Nd systematics supported by the unexpectedly young ^{146}Sm-^{142}Nd "age" of this meteorite in comparison with its otherwise good behavior for most other radiometric systems"* (R.W. Carlson, written communication, 2020)

can be viewed as a general question if the entirety of its consituent mineral phases are pristine. If even in the achondrite D'Orbigny the mineral phases that provide robust radiometric information for the Mn-Cr system (Glavin et al., 2004) coexist with mineral phases carrying a disturbed Sm-Nd system, it becomes difficult to argue "beyond reasonable doubt" that such disturbances of the Sm-Nd system were absent in meteorites whose other radiometric systems are not pristine. In the search for a "point-like" calibrator for the ^{146}Sm half-life, it must be explicitly stressed that the default assumption must never be that a given mineral chronometer behaves ideally. On the contrary, the burden of proof must lie on the proponent of a calibrator, who needs to actively demonstrate the degree of ideality of the proposed chronometric system.

The open-system behavior documented by Sanborn et al. (2015) manifests itself in a variability of the subjectively chosen ages for at least two of the four lunar samples analyzed by the Carnegie-Livermore collaboration. If one uses the ^{147}Sm-^{143}Nd age for lunar rock 77215 determined by Carlson et al. (2014), 4.42 ± 0.07 Ga, instead of that by Marks et al. (2014), 4.29 Ga, the resulting ^{146}Sm/^{147}Sm ratio is compatible with the 68 Ma half-life but not the 103 Ma half-life for ^{146}Sm. Troctolite 76535 (for which a dozen mutually incompatible ages have been produced over the years), has a ^{207}Pb/^{206}Pb age of 4.38 Ga, the Rb-Sr age with the Villa et al. (2015) half-life of ^{87}Rb is 4.33 ± 0.05 Ga, and the ^{146}Sm-^{142}Nd isochron translates to an age of 4.38 ± 0.02 Ga when assuming a ^{146}Sm half-life of 68 Ma.

Both the paper by Carlson et al. (2014) and the abstracts cited by Marks et al. (2014) often calculate internal isochrons that exclude the fine sieve fraction due to its isotopic disequilibrium with the other minerals. While excluding a particular sieve fraction can produce an approximate age that is fit for the intended purpose of these papers, namely estimating the age of lunar magma ocean formation, the fact remains that the sample is in petrological disequilibrium. In this case, it is not correct to assume that sieving can get rid of all alteration products. Alteration occurs at the sub-µm scale and it is expected that even the coarse grains selected by handpicking do contain some alteration products, as indeed shown by the overdispersion of the calculated isochrons and acknowledged in the verbatim quote from the Carlson at al. (2014) paper cited above. This means that none of the lunar rocks in Fig. 4 of Marks et al. (2014) is fit-for-use as a standard reference material on which to pin the calibration of the ^{146}Sm half-life.

Figure Captions

Figure 1 - ^{147}Sm half-life counting measurements of the last 60 years, as reported in Table 1. The red dots represent the measured values, and the vertical blue line segments depict the measured values plus or minus one associated standard uncertainty. The horizontal dark green line indicates the consensus value, and the height of the light green rectangle centered on it represents the consensus value plus or minus one associated standard uncertainty.

Figure 2 - All ^{146}Sm half-life counting measurements, as reported in Table 3. The red dots represent the measured values, and the vertical blue line segments depict the measured values plus or minus one associated standard uncertainty.