

Determination of the Radioisotope Decay Constants and Half-Lives: Samarium-147 (^{147}Sm)

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Abstract

Over the last 80 years numerous determinations have been made of the ^{147}Sm half-life. The determinations since 1960 have converged on close agreement between the two primary determination techniques used in direct physical counting experiments—ionization chambers and liquid scintillation counters, and with radioisotope age comparisons using two meteorites. Thus the ^{147}Sm half-life value of $106 \pm 0.8 \text{ Byr}$ has now been adopted for standard use by the uniformitarian geological community. This value is based on the weighted average of four direct counting determinations in the period 1961–1970 and the recalibration in the 1970s of Sm-Nd model ages of two meteorites by forcing them (essentially by circular reasoning) to agree with their Pb-Pb isochron and model ages. However, direct counting experiments in 2003 determined the ^{147}Sm half-life value was 10% or more longer at $117 \pm 2 \text{ Byr}$. This was achieved by using four standard Sm solutions with internal α -radioactive standards in 19 alpha spectrometer and 24 ionization chamber determinations, making it the most thorough and comprehensive effort to determine the ^{147}Sm half-life. The thinner counting sources used, while resulting in low α -activities being measured, greatly reduced the counting uncertainty due to self-absorption of the emitted α -particles. Although rejected or ignored, this $117 \pm 2 \text{ Byr}$ value for the ^{147}Sm half-life, which agrees with some earlier determinations, may well be highly significant and more reliable than the adopted value. Yet, in spite of the many experiments directly measuring ^{147}Sm decay, preference has been given to the half-life value of $106 \pm 0.8 \text{ Byr}$ determined by forcing the Sm-Nd data to agree with Pb-Pb dates. But many unprovable assumptions are also involved, not the least being that the radioisotope systems closed at the same time and subsequently remained closed. Furthermore, even this "gold standard" has unresolved uncertainties due to the U decay constants being imprecisely known, and to measured variations of the $^{238}\text{U}/^{235}\text{U}$ ratio in terrestrial rocks and minerals and in meteorites. Both of these factors are so critical to the U-Pb method, as well as the additional factor of knowing the initial concentrations of the daughter and index isotopes, so it should not be used as a standard to determine other decay constants. In any case, the determined half-life of ^{147}Sm has been shown to be dependent on the thicknesses of the Sm counting source and the detector. There is also evidence decay rates of the radioisotopes used for rock dating have not been constant in the past. This only serves to emphasize that if the Sm-Nd dating method has been calibrated against the U-Pb "gold standard" with all its attendant uncertainties, then it cannot be absolute, and therefore it cannot be used to reject the young-earth creationist timescale. Indeed, current radioisotope dating methodologies are at best hypotheses based on extrapolating current measurements and observations back into an assumed deep time history for the cosmos.

Keywords: radioisotope dating, decay constants, half-lives, samarium-147, ^{147}Sm , α decay, direct counting, emulsions, liquid scintillation counters, ionization chambers, surface-barrier detectors, Geiger counter, counting efficiencies, geological comparisons, meteorites, U-Pb "gold standard", $^{238}\text{U}/^{235}\text{U}$ ratio

Introduction

Radioisotope dating of rocks and meteorites is perhaps the most potent claimed proof for the supposed old age of the earth and the solar system. The absolute ages provided by the radioisotope dating methods provide an apparent aura of certainty to the claimed millions and billions of years for formation of the earth's rocks. Many in both the scientific community and the general public around the world thus remain convinced of the earth's claimed great antiquity.

However, accurate radioisotopic age determinations require that the decay constants of the respective parent radionuclides be accurately known and constant in time. Ideally, the uncertainty

of the decay constants should be negligible compared to, or at least be commensurate with, the analytical uncertainties of the mass spectrometer measurements of isotopic ratios entering the radioisotope age calculations (Begemann et al. 2001). Clearly, based on the ongoing discussion in the conventional literature this is not the case at present. The stunning improvements in the performance of mass spectrometers during the past four or so decades, starting with the landmark paper by Wasserburg et al. (1969), have not been accompanied by any comparable improvement in the accuracy of the decay constants (Begemann et al. 2001; Steiger and Jäger 1977), in spite of ongoing attempts (Miller 2012). The uncertainties associated with most direct

half-life determinations are, in most cases, still at the 1% level, which is still significantly better than any radioisotope method for determining the ages of rock formations. However, even uncertainties of only 1% in the half-lives lead to very significant discrepancies in the derived radioisotope ages. The recognition of an urgent need to improve the situation is not new (for example, Min et al. 2000; Renne, Karner, and Ludwig 1998). It continues to be mentioned, at one time or another, by every group active in geochronology (Schmitz 2012). This is a key issue especially for very long half-life radioisotopes due to the very slow accumulation of decay particle counting data, because the statistical error is equal to the square root of the total decay particle counts.

From a creationist perspective, the 1997–2005 RATE (Radioisotopes and the Age of The Earth) project successfully made progress in documenting some of the pitfalls in the radioisotope dating methods, and especially in demonstrating that radioisotope decay rates may not have always been constant at today's measured rates (Vardiman, Snelling, and Chaffin 2000, 2005). Yet much research effort remains to be done to make further inroads into not only uncovering the flaws intrinsic to these long-age dating methods, but towards a thorough understanding of radioisotopes and their decay during the earth's history within a biblical creationist framework.

One crucial area the RATE project did not touch on was the issue of how reliable have been the determinations of the radioisotope decay rates, which are so crucial for calibrating these dating "clocks." Indeed, before this present series of papers (Snelling 2014a, 2014b, 2015) there have not been any attempts in the creationist literature to review how the half-lives of the parent radioisotopes used in long-age geological dating have been determined and to collate their determinations so as to discuss the accuracy of their currently accepted values. After all, accurate radioisotope age determinations depend on accurate determinations of the decay constants or half-lives of the respective parent radioisotopes. The reliability of the other two assumptions these absolute dating methods rely on, that is, the starting conditions and no contamination of closed systems, are unprovable. Yet these can supposedly be circumvented somewhat via the isochron technique, because it is independent of the starting conditions and is sensitive to revealing any contamination, which is still significantly better than any radioisotope method for determining the ages of rock formations. Data points that do not fit on the isochron are simply ignored because their values are regarded as due to contamination. Yet there is also no reliable way of determining the difference between isochrons and mixing lines. That

this is common practice is illustrated with numerous examples from the literature by Dickin (2005) and Faure and Mensing (2005). On the other hand, it could be argued that this discarding of data points which do not fit the isochron is arbitrary and therefore is not good science, because it is merely assumed the "aberrant" values are due to contamination rather than that being proven to be so. Indeed, in order to discard such outliers in any data set, one must establish a reason for discarding those data points which cannot be reasonably questioned.

In order to rectify this deficiency, Snelling (2014a, 2014b, 2015) has documented the methodology behind and history of determining the decay constants and half-lives of the parent radioisotopes ^{87}Rb , ^{176}Lu , and ^{187}Re used as the basis for the Rb-Sr, Lu-Hf, and Re-Os long-age dating methods respectively. He showed that there is still some uncertainty in what the values for these measures of the ^{87}Rb and ^{176}Lu decay rates should be, in contrast to the apparent agreement on the ^{187}Re decay rate. This uncertainty is especially prominent in determinations of the ^{176}Lu decay rate by physical direct counting experiments. Furthermore, the determined values of the ^{87}Rb decay rate differ when Rb-Sr ages are calibrated against the U-Pb ages of either the same terrestrial minerals and rocks or the same meteorites and lunar rocks. Ironically it is the slow decay rate of isotopes such as ^{87}Rb used for deep time dating that makes a precise measurement of that decay rate so difficult. Thus it could be argued that direct measurements of these decay rates should be the only acceptable experimental evidence, especially because measurements which are calibrated against other radioisotope systems are already biased by the currently accepted methodology that the secular community uses in their rock dating methods. Indeed, the ^{87}Rb , ^{176}Lu , and ^{187}Re decay half-lives have all ultimately been calibrated against the U-Pb radioisotope system, yet there are now known measured variations in the $^{238}\text{U}/^{235}\text{U}$ ratio that is critical to that method (Brennecka and Wadhwa 2012; Hiess et al. 2012).

Therefore, the aim of this contribution is to further document the methodology behind and history of determining the present decay constants and half-lives of the parent radioisotopes used as the basis for the long-age dating methods. It is necessary to explore just how accurate these determinations are, whether there really is consensus on standard values for the half-lives and decay constants, and just how independent and objective the standard values are from one another between the different methods. Of course, it is to be expected that every long-lived radioactive isotope is likely to show similar variation and uncertainty in half-life measurements

because these are difficult measurements to make. However, even small variations and uncertainties in the half-life values result in large variations and uncertainties in the calculated ages for rocks, and the question remains as to whether the half-life values for each long-lived parent radioisotope are independently determined. We continue here with samarium-147 (¹⁴⁷Sm), which is the basis for the Sm-Nd dating method.

Samarium and Samarium-147 Decay

Samarium (Sm) and neodymium (Nd) are both rare-earth elements (REEs) with atomic numbers (*Z*) of 62 and 60 respectively. The rare-earth elements generally form ions with a 3+ charge whose radii decrease with increasing atomic number from 1.15 Å in lanthanum (La), atomic number 57, to 0.93 Å in lutetium (Lu), atomic number 71 (Faure and Mensing 2005). The REEs occur in high concentrations in several economically important minerals such as bastnaesite (CeFCO₃), monazite (CePO₄), and cerite [(Ca,Mg)₂(Ce)₈(SiO₄)₇·3H₂O]. Furthermore, they occur as trace elements in common rock-forming minerals (silicates, phosphates, and carbonates) in which they replace major element ions. They may also reside in inclusions of certain accessory minerals in the common rock-forming silicates.

Minerals exercise a considerable degree of selectivity in admitting REEs into their crystal structures (Faure and Mensing 2005). Feldspar, biotite, and apatite tend to concentrate the light REEs (the Ce group), whereas pyroxenes, amphiboles, and garnet concentrate the heavy REEs (the Gd group). The selectivity of the rock-forming minerals for the light or heavy REEs obviously affects the REE concentrations of the rocks in which those minerals occur. Sm and Nd both belong to the light REEs, so they tend to concentrate in feldspar, biotite, and apatite. Thus the Sm and Nd concentrations in calc-alkaline plutonic and volcanic igneous rocks range from <1 ppm in ultramafic rocks to about 8 ppm Sm and 45 ppm Nd in granite. Alkali-rich igneous rocks have consistently higher Sm and Nd concentrations than the calc-alkaline suite, ranging up to about 15 ppm Sm and 85 ppm Nd.

Sm and Nd exhibit an unusual geochemical behavior, which arises from what is known as the “lanthanide contraction” (Faure and Mensing 2005). This contraction results from the way electrons fill their *f* shell orbitals. As a consequence, the ionic radius of Sm (*Z*=62) is smaller than that of Nd (*Z*=60). Even though the difference in the radii is small (Nd³⁺=1.08 Å; Sm³⁺=1.04 Å), Nd is preferentially concentrated in the liquid phase during partial melting of silicate minerals, whereas Sm remains in the residual solids. For this reason, basalt

magma have lower Sm/Nd ratios than the source rocks from which they formed. Thus this preferential partitioning of Nd into the melt phase has caused the rocks of the continental crust to be enriched in Nd relative to Sm compared to the residual rocks in the lithospheric mantle.

Even though the concentrations of Sm and Nd reach high values in accessory phosphate minerals such as apatite and monazite and in carbonatites, these minerals and carbonatites are still more enriched in Nd than in Sm and hence their Sm/Nd ratios are less than 0.32. Among the rock-forming silicate minerals, garnet is the only one with a high Sm/Nd ratio (0.54) even though its concentrations of Sm and Nd are both low (1–2 ppm). Several other rock-forming silicate minerals, such as K-feldspar, biotite, amphibole, and clinopyroxene, have higher Sm and Nd concentrations than garnet, but their Sm/Nd ratios are less than 0.32 in most cases.

Sm and Nd each have seven naturally-occurring isotopes. Of these ¹⁴⁷Sm, ¹⁴⁸Sm, and ¹⁴⁹Sm are all radioactive, but the latter two have such long half-lives (about 10¹⁶ years) that they are not capable of producing measurable variations in the daughter isotopes ¹⁴⁴Nd and ¹⁴⁵Nd, even over supposed conventional cosmological intervals (10¹⁰ years) (Dickin 2005). Yet ¹⁴⁷Sm only has an abundance of 15.0% in naturally occurring Sm (Lide and Frederikse 1995). Although the half-life of ¹⁴⁷Sm is also very long (currently determined as 106 billion years), it decays by α-particle emission to ¹⁴³Nd, a stable isotope of Nd. The relevant decay scheme is often depicted as:



where ⁴He is an α-particle and *E* is the total decay energy. The energy of the ¹⁴⁷Sm emitted α-particles is 2.23 MeV. This decay scheme has proven useful to uniformitarians for apparently dating terrestrial rocks, stony meteorites (both chondrites and achondrites), and lunar rocks.

There are two parameters by which the decay rate is measured and expressed, namely, the decay constant (λ) and the half-life ($t_{1/2}$). The decay constant can be defined as the probability per unit time of a particular nucleus decaying, though strictly speaking probabilities do not have units associated with them and the decay constant is derived from a definitive functional relationship. In contrast, the half-life is the time it takes for half of a given number of the parent radionuclide atoms to decay. The two quantities can be almost used interchangeably, because they are related by the equation:-

$$t_{1/2} = \ln 2/\lambda = 0.693/\lambda$$

The decay rate of ¹⁴⁷Sm has not been all that difficult to determine once the necessary instrumentation

was developed to accurately count the emitted α -particles. However, the Sm-Nd dating method has had its problems.

The lanthanide contraction causes the distribution of Sm and Nd to be opposite to that of Rb and Sr (Faure and Mensing 2005). And because Sm and Nd have very similar chemical properties (unlike Rb and Sr), large ranges of Sm/Nd ratios in whole-rock systems are rare, and in particular low Sm/Nd ratios near the vertical y-axis on an isochron dating graph are very rare. Therefore, because of the difficulty of obtaining a wide range of Sm/Nd ratios from a single rock body, and because of the greater technical demands of Nd isotope analysis, the Sm-Nd isochron dating method has been generally only applied to dating rock units for which Rb-Sr isochron dating has proven unsatisfactory. Many of those applications were also made before the U-Pb zircon dating method had reached its present level of development. Therefore, some of those rock units have subsequently been dated to apparent greater accuracy and precision by the U-Pb method. Nevertheless, the Sm-Nd method has continued to be used to date rocks and meteorites and thus the determination of the ^{147}Sm half-life used by the method requires examination.

Determination Methods

Attempts to measure the α -radioactivity of Sm date back to the early 1930s. Famous names, such as Hevesy and Pahl, Curie and Joliot, Libby, and many more, are among the researchers who studied this phenomenon by various techniques (Begemann et al. 2001). At that time, before ^{147}Sm had finally been identified as the isotope accountable for the α -radioactivity of Sm (Weaver 1950), the half-life was calculated in terms of the total element of Sm, with results ranging from 0.63 to 1.4×10^{12} years (that is, 630 to 1400 Byr). Even in 1949, when ^{148}Sm (Wilkins and Dempster 1938) and ^{152}Sm (Dempster 1948) had been reported erroneously to be responsible for the α -activity of samarium, Picciotto still published his result in terms of total Sm as $6.7 \pm 0.4 \times 10^{11}$ years (670 Byr), this quoted statistical error amounting to approximately 300 observed decays of ^{147}Sm . Almost all attempts to measure the α -radioactivity of ^{147}Sm have been by direct counting of α -particles, although two geological comparisons of radioisotope ages of individual meteorites have been used to confirm the direct counting measurements of the ^{147}Sm half-life.

Direct counting

Initially direct measurements were done by counting α -particles registered on a photographic emulsion. For example, Picciotto (1949) used solutions of samarium sulfate to deposit layers of calculated weights of samarium across photographic

emulsions which were then left exposed to the samarium α -activity for four to more than 19 days. At the end of each exposure time interval the tracks left by the α -particles on the emulsions when developed were counted. The calculated numbers of α -particles per second per gram of Sm were averaged to derive a decay rate in terms of the total Sm (all isotopes) of $6.7 \pm 0.4 \times 10^{11}$ years (670 Byr). Even though Beard and Wiedenbeck (1954) obtained a ^{147}Sm half-life value using an ionization chamber (Geiger counter), they corroborated their result by checking the energy spectrum of their samarium source by exposing it for three weeks to a nuclear plate, which registered the tracks left by the α -particles. The mean track length compared favorably with previous emulsion experiments, and the energy distribution confirmed the tracks were produced by α -particles emitted by ^{147}Sm .

A more recent use of the emulsion method by Martins, Terranova, and Moreira Correa (1992) involved spreading a calibrated solution of samarium nitrate over a glass plate which was then heated to leave a stable film of samarium oxide whose uniform thickness was measured. This method enabled accurate control, within the limits of volumetric error, of the quantity of samarium deposited on the glass plate. This glass plate was then contacted against a plastic emulsion plate for registration of the spontaneous ^{147}Sm α -particle emission. After exposure for 30 days, stored in an underground laboratory to protect it from cosmic radiation, the emulsion was processed to etch the tracks left by the α -particles, which were then counted. The samarium oxide film was also checked by γ -spectrometry to rule out any contribution to the counted tracks from the possible presence of any U or Th atoms. A measurement of the ^{147}Sm half-life was then calculated from the number of α -particle tracks produced from the known quantity of samarium in the exposure time period of 30 days. Though they did not account for possible α -particle energy loss in the samarium or the glass and plastic plates, this possibility was deemed negligible.

However, since the mid-1950s a variety of instruments have primarily been used for direct counting of ^{147}Sm α -particles. In these instruments, the alpha (α) activity of ^{147}Sm in a source material is counted over a designated time period, and divided by the total number of radioactive ^{147}Sm atoms in the known quantity of Sm, based on Avogadro's number and the isotopic abundance of ^{147}Sm . Two types of such instruments have been used to count the α -particles emitted from different ^{147}Sm sources—liquid scintillation spectrometers (Beard and Kelly 1958; Donhoffer 1964; Kinoshita, Yokoyama, and Nakanashi 2003; Kossert et al. 2009; Wright, Steinberg, and Glendenin 1961), and

ionization chambers or Geiger counters (Beard and Wiedenbeck 1954; Gupta and MacFarlane 1970; Karras and Nurmia 1960; Kinoshita, Yokoyama, and Nakanashi 2003; MacFarlane and Kohman 1961). A variety of ^{147}Sm sources have also been used, namely, samarium octoate (Wright, Steinberg, and Glendenin 1961), samarium oxide (Beard and Wiedenbeck 1954; Kinoshita, Yokoyama, and Nakanashi 2003; Kossert et al. 2009; MacFarlane and Kohman 1961; Su et al. 2010), and samarium metal (Gupta and MacFarlane 1970; Su et al. 2010).

In the most recent half-life determination, Su et al. (2010) deposited ^{147}Sm -enriched metal and oxide on pure quartz glass substrates by vacuum evaporation and sputtering respectively, and then checked the uniformity of the thicknesses of these samples by exposing them to plastic emulsion plates in direct contact with them. However, in using this method it is difficult to compensate for α -energy loss due to absorption. After 100 hours exposure the emulsion plates were chemically etched and the α -particle tracks were observed and counted. This does not appear to be a good way of establishing uniformity of thickness, whereas α -gauging would be a better method. Nevertheless, the α -activities of the two samples were then measured by silicon surface-barrier detectors placed in vacuum chambers for a period of 200 hours (8 days 8 hours). Finally the ^{147}Sm half-life was then calculated from the α -activity spectra for each sample (Sm metal and Sm oxide).

A liquid scintillation counter or spectrometer detects and measures ionizing radiation by using the excitation effect of incident α -particles on a scintillator material, and detecting the resultant light pulses. It consists of a scintillator which generates photons of light in response to incident α -particles, a sensitive photomultiplier tube which converts the light to an electrical signal, and electronics to process this signal.

Liquid scintillation counting measures the α -activity of a sample, prepared by mixing the α -active material with a liquid scintillator, and counting the resultant photon emissions. This allows for more efficient counting due to the intimate contact of the α -activity with the scintillator. Samples are dissolved or suspended in a "cocktail" containing a solvent, typically some form of a surfactant, and small amounts of other additives known as "fluors" or scintillators.

The radioactive sample is then placed in a vial containing a premeasured amount of scintillator cocktail and this vial plus vials containing known amounts of ^{147}Sm are loaded into the liquid scintillation counter. Many counters have two photomultiplier tubes connected in a coincidence circuit. The coincidence circuit assures that genuine light pulses, which reach both photomultiplier tubes,

are counted, while spurious pulses (due to line noise, for example), which would only affect one of the tubes, are ignored.

When a charged particle strikes the scintillator, its atoms are excited and photons are emitted. These are directed at the photomultiplier tube's photocathode, which emits electrons by the photoelectric effect. These electrons are electrostatically accelerated and focused by an electrical potential so that they strike the first dynode of the tube. The impact of a single electron on the dynode releases a number of secondary electrons which are in turn accelerated to strike the second dynode. Each subsequent dynode impact releases further electrons, and so there is a current amplifying effect at each dynode stage. Each stage is at a higher potential than the previous stage to provide the accelerating field. The resultant output signal at the anode is in the form of a measurable pulse for each photon detected at the photocathode, and is passed to the processing electronics. The pulse carries information about the energy of the original incident α -radiation on the scintillator. Thus both the intensity and energy of the α -particles can be measured.

The scintillation spectrometer consists of a suitable scintillator crystal, a photomultiplier tube, and a circuit for measuring the height of the pulses produced by the photomultiplier. The pulses are counted and sorted by their height, producing an x-y plot of scintillator flash brightness versus number of flashes, which approximates the energy spectrum of the incident α -radiation.

The ionization chamber is the simplest of all gas-filled radiation detectors, and is widely used for the detection and measurement of certain types of ionizing radiation, including α -particles (fig. 1). Conventionally, the term "ionization chamber" is used exclusively to describe those detectors which collect all the charges created by direct ionization within the gas through the application of an electric field. It only uses the discrete charges created by each interaction between the incident radiation and the gas, and does not involve the gas multiplication mechanisms used by other radiation instruments, such as the Geiger-Müller counter or the proportional counter.

An ionization chamber measures the current from the number of ion pairs created within a gas caused by incident α -radiation. It consists of a gas-filled chamber with two electrodes, known as the anode and cathode (fig. 1). The electrodes may be in the form of parallel plates, or a cylinder arrangement with a coaxially located internal anode wire. A voltage is applied between the electrodes to create an electric field in the fill gas. When gas between the electrodes is ionized by incident ionizing α -radiation (or β - or γ -radiation), ion-pairs are created and the resultant positive ions

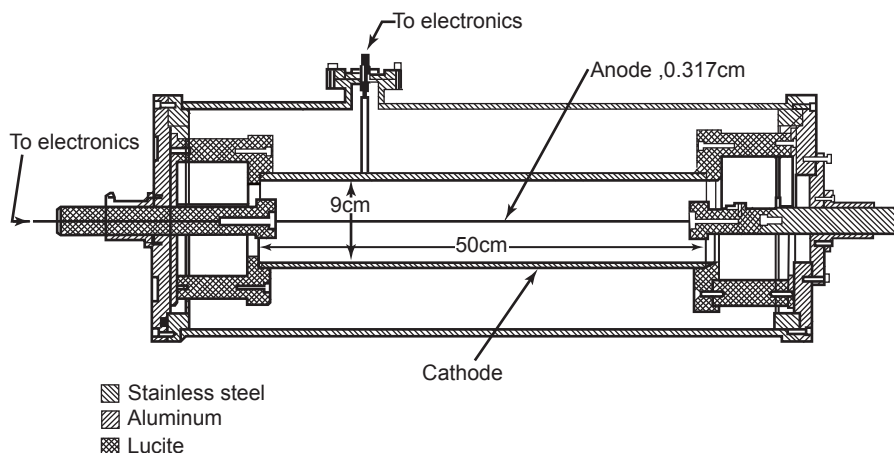


Fig. 1. Schematic diagram of the cylindrical ionization chamber (alpha spectrometer) used by Gupta and MacFarlane (1970) in their determination of the ^{147}Sm half-life.

and dissociated electrons move to the electrodes of the opposite polarity under the influence of the electric field. This generates an ionization current which is measured by an electrometer. The electrometer must be capable of measuring the very small output current which is in the region of femtoamperes (10^{-15} amps) to picoamperes (10^{-12} amps), depending on the chamber design, α -radiation dose and applied voltage. The unique feature of ionization chambers is that the electric field strength is low enough that no multiplication of ion pairs occurs. Hence the current generated at a given voltage depends on the type and energy of the incident radiation but is independent over a range of applied voltages, approximately 100–300 volts.

Each ion pair creates deposits or removes a small electric charge to or from an electrode, such that the accumulated charge is proportional to the number of ion pairs created, and hence the α -radiation dose. This continual generation of charge produces an ionization current, which is a measure of the total ionizing dose entering the chamber. The electric field also enables the device to work continuously by mopping up electrons, which prevents the fill gas from becoming saturated, where no more ions could be collected, and by preventing the recombination of ion pairs, which would diminish the ion current. This mode of operation is referred to as “current” mode, meaning that the output signal is a continuous current, and not a pulse output as in the cases of the Geiger-Müller tube or the proportional counter. In the ionization chamber operating region the collection of ion pairs is effectively constant over a range of applied voltage, as due to its relatively low electric field strength the ion chamber does not have any multiplication effect. This is in distinction to the Geiger-Müller tube or the proportional counter whereby secondary electrons, and ultimately multiple avalanches, greatly amplify the original

ion-current charge. In the proportional counter the electric field produces discrete, controlled avalanches such that the energy and type of radiation can be determined at a given applied field strength.

The Geiger-Müller counter, also called a Geiger counter, is also used for measuring ionizing radiation. It detects radiation such as α -particles, using the ionization produced in a Geiger-Müller tube. The processing electronics displays the result. The Geiger-Müller tube is filled with an inert gas such as helium, neon, or argon at low pressure, to which a high voltage is applied. The tube briefly conducts an electrical charge when a particle or photon of incident α -radiation makes the gas conductive by ionization. The ionization is considerably amplified within the tube by an avalanche effect to produce an easily measured detection pulse, which is fed to the processing and display electronics. The electronics also generates the high voltage, typically 1000–1400 volts, which has to be applied to the Geiger-Müller tube to enable its operation. The voltage must be high enough to produce avalanche effects for all incident radiation. Thus a Geiger-Müller tube has no ability to discriminate between incident radiations; all radiation produces the same current.

There are two main limitations of the Geiger counter. Because the output pulse from a Geiger-Müller tube is always the same magnitude regardless of the energy of the incident α -radiation, the tube cannot differentiate between radiation types. A further limitation is the inability to measure high α -radiation intensities due to the “dead time” of the tube. This is an insensitive period after each ionization of the gas during which any further incident α -radiation will not result in a count, and the indicated rate is therefore lower than actual. Typically the dead time will reduce indicated count rates above about 10^4 to 10^5 counts per second depending on the characteristic of the tube being

used. Whilst some counters have circuitry which can compensate for this, for accurate measurements ion chamber instruments are preferred to measure high radiation rates.

Counting efficiencies of liquid scintillation counters under ideal conditions range from about 30% for tritium (a low-energy β -emitter), to nearly 100% for phosphorus-32 (^{32}P), a high-energy β -emitter. Thus counting efficiencies for α -particles are within this range, which is <100%. Some chemical compounds (notably chlorine compounds) and highly colored samples can interfere with the counting process. This interference, known as quenching, can be overcome through data correction or through careful sample preparation. In ionization counters the counting efficiency is the ratio between the number of α -particles or photons counted and the number of α -particles or photons of the same type and energy emitted by the α -radiation source. Counting efficiencies vary for different isotopes and sample compositions, and for different scintillation counters. Poor counting efficiency can be caused by an extremely low energy to light conversion rate (the scintillation efficiency), which, even optimally, will be a small value. It has been calculated that only some 4% of the energy from a β -emission event is converted to light by even the most efficient scintillation cocktails. Proportional counters and end-window Geiger-Müller tubes have a very high efficiency for all ionizing particles that reach the fill gas. Nearly every initial ionizing event in the gas will result in avalanches, and thereby an output signal. However the overall detector efficiency is largely affected by attenuation due to the window or tube body through which particles have to pass. They are also extremely sensitive to various types of background radiation due to their lack of discrimination.

Judged from the fact that many of these direct counting experiments, particularly the earlier ones, have yielded results that are not compatible with one another within the stated uncertainties (see below), it would appear that not all the measurement uncertainties are accounted for, and therefore the stated uncertainties are likely unrealistically small and typically are underestimated. Begemann et al. (2001) maintain that many of such experiments are likely plagued by unrecognized systematic errors. As the nature of these errors is obscure, it is not straightforward to decide which of the, often mutually exclusive, results of such direct counting experiments is closest to the true value, although most of the post-early-1960s experiments appear to converge on a common value (see below). Furthermore, the presence of unknown systematic biases makes any averaging dangerous. It is possible that reliable results of

careful workers, listing realistic uncertainties, will not be given the weights they deserve—this aside from the question of whether it makes sense to average numbers that by far do not all agree within the stated uncertainties.

Geological comparisons of methods

A second approach used by secular scientists to determine the ^{147}Sm decay half-life has been to date geological samples whose ages have also been measured by other methods with presumably more reliable decay constants (Dickin 2005; Faure and Mensing 2005). This essentially involves circular reasoning, because it is being assumed the other radioisotope dating methods, principally the U-Pb method, gives the reliable dates to which the ^{147}Sm half-life can be calibrated to bring the Sm-Nd radioisotope ages into agreement. It should be noted, however, that this is hardly objective, because all the radioisotope ages of rocks could be wrong due to the underlying unprovable and suspect assumptions on which all the radioisotope dating methods are based. Nevertheless, a few geological determinations of the ^{147}Sm half-life were made in the 1970s using components of individual achondrite meteorites (Dickin 2005, 70–71; Lugmair 1974; Lugmair, Scheinin, and Marti 1975; Lugmair and Marti 1977).

This method has the disadvantage that it involves geological uncertainties, such as whether all isotopic systems closed at the same time and remained closed. However, it is claimed to still provide a useful check on the laboratory determinations by direct physical counting. Nevertheless, this approach entails multi-chronometric dating of minerals and components in individual meteorites and cross-calibration of different radioisotopic age systems by adjusting the decay constant of the Sm-Nd system so as to force agreement with the age obtained via another dating system, usually U-Pb (Begemann et al. 2001). In essence, because the half-life of ^{238}U is claimed to be the most accurately known of all relevant radionuclides, this usually amounts to expressing ages in units of the half-life of ^{238}U .

Results of the Samarium-147 Decay Determinations

During the last 80 years numerous determinations of the ^{147}Sm decay constant and half-life have been made using these methods. The results are listed with details in Table 1. The year of the determination versus the value of the half-life is plotted in Fig. 2. In each case the data points plotted have been color-coded the same to differentiate the values as determined by the two approaches that have been used—direct counting, and geological comparisons with other radioisotope dating methods.

Discussion

Results obtained after 1954, and particularly during the 1960s and the ensuing decade, began to converge towards a ^{147}Sm common half-life value (see table 1 and fig. 2). In the early 1970s, when Lugmair and his colleagues began to develop the decay of ^{147}Sm to ^{143}Nd as a dating tool (Lugmair 1974), they used only a weighted average of the last four half-life measurements at that time,

those of Wright, Steinberg, and Glendenin (1961), Donhoffer (1964), Valli et al. (1965), and Gupta and MacFarlane (1970) (Lugmair and Marti 1978). They range from 1.04 to 1.08×10^{11} years (104 to 108 Byr) with a weighted mean of $1.060 \pm 0.008 \times 10^{11}$ years (106 ± 0.8 Byr) (1 σ uncertainty). It is worth noting that the statistical error of this weighted mean infers that more than 10,000 α -decays of ^{147}Sm have occurred to produce this result. Nevertheless, it

Table 1. Determinations of the ^{147}Sm decay rate expressed in terms of the half-life using direct physical counting experiments, and comparisons of radioisotope ages of terrestrial minerals and rocks, and meteorites.

Determination of the Sm Decay Rate						
Date	Half-Life (Byr)	Uncertainty (Byr)	Method	Instrument	Notes	Source
1936	150	± 11	direct counting	emulsion plate		Hosemann 1936
1949	100	± 6	direct counting	emulsion plate		Picciotto 1949
1954	125	± 6	direct counting	ionization chamber	4 π Geiger counter	Beard and Wiedenbeck 1954
1958	128	± 4	direct counting	liquid scintillation		Beard and Kelly 1958
1960	114	± 5	direct counting	ionization chamber		Karras and Nurmia 1960
1960	117	± 5	direct counting	ionization chamber		Karras 1960
1961	113		direct counting			Graeffe and Nurmia 1961
1961	115	± 5	direct counting	ionization chamber		MacFarlane and Kohman 1961
1961	105	± 1	direct counting	liquid scintillation		Wright, Steinberg, and Glendenin 1961
1964	104	± 3	direct counting	liquid scintillation		Donhoffer 1964
1965	108	± 2	direct counting	liquid scintillation		Valli et al. 1965
1970	106	± 2	direct counting	ionization chamber		Gupta and MacFarlane 1970
1975	106		geological comparisons		Juvinas basaltic achondrite meteorite	Lugmair, Sheinin, and Marti 1975
1977	106		geological comparisons		Angra dos Reis achondrite meteorite	Lugmair and Marti 1977
1978	106	± 0.8	direct counting		weighted average of four measures	Lugmair and Marti 1978
1987	105	± 4	direct counting			Al-Bataina and Jänecke 1987
1992	123	± 4	direct counting	emulsion plate		Martins, Terranova, and Moreira Correa 1992
2001	106	± 4	direct counting	emulsion plate	correction of Martins et al. 1992	Begemann et al. 2001
2003	117	± 2	direct counting	alpha spectrometer	with vacuum chamber (also liquid scintillation)	Kinoshita, Yokoyama, and Nakanashi 2003
2009	107	± 0.9	direct counting	liquid scintillation		Kossert et al. 2009
2010	106	± 1	direct counting	alpha spectrometer	with vacuum chamber, Sm metal	Su et al. 2010
2010	107	± 1	direct counting	alpha spectrometer	with vacuum chamber, Sm oxide	Su et al. 2010

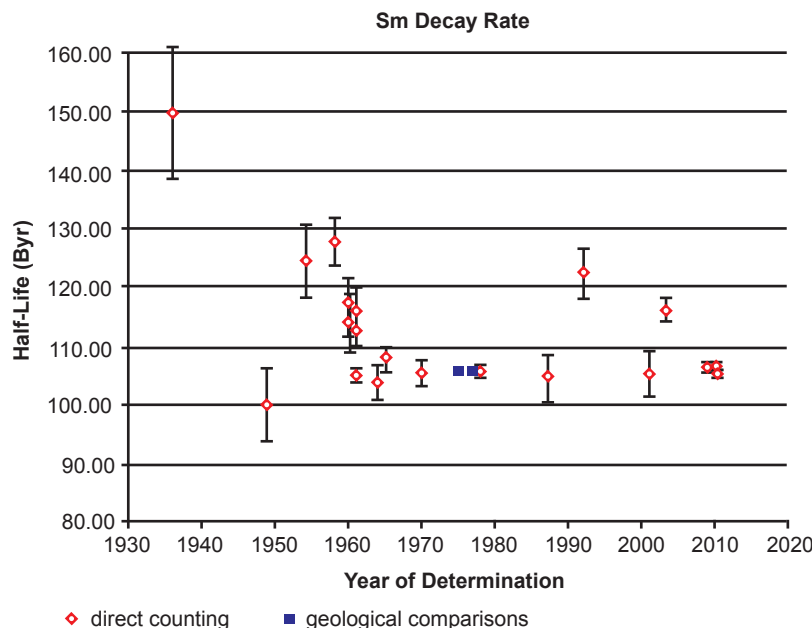


Fig. 2. Plot of each ^{147}Sm half-life determination versus the year of its determination, color-coded according to the method of its determination. The error bars for each determination are also plotted from the error values listed in Table 1.

should be noted that three of those measurements were made using liquid scintillation counters and one using an ionization chamber (see table 1), all of which have different counting efficiencies, as already discussed. Nevertheless, that ^{147}Sm half-life value has been adopted by all geochronologists and cosmochronologists since that time (the late 1970s).

There have been five more modern measurements of the ^{147}Sm half-life since the 1970s (table 1). The first by Al-Bataina and Jänecke (1987) with a value of $1.05 \pm 0.04 \times 10^{11}$ years (105 ± 4 Byr) agrees very well with the previous direct counting results of Donhoffer (1964), Gupta and MacFarlane (1970), Valli et al. (1965), and Wright, Steinberg, and Glendenin (1961), as well as with the geological comparisons with Pb-Pb ages of meteorites by Lugmair (1974), Lugmair, Scheinin, and Marti (1975), and Lugmair and Marti (1977) (table 1). It is also in close agreement with subsequent determinations by Kossert et al. (2009) and Su et al. (2010) (table 1). However, the determination by Martins, Terranova, and Moreira Correa (1992) of $1.23 \pm 0.04 \times 10^{11}$ years (123 ± 4 Byr) is substantially higher than the Al-Bataina and Jänecke (1987) determination of $1.05 \pm 0.04 \times 10^{11}$ years (105 ± 4 Byr), as is the Kinoshita, Yokoyama, and Nakanashi (2003) determination of $1.17 \pm 0.02 \times 10^{11}$ years (117 ± 2 Byr) (table 1). These variant results can be easily seen in Fig. 2.

Begemann et al. (2001) claimed that because there is good agreement of ages obtained using the generally accepted ^{147}Sm half-life value of $1.06 \pm 0.01 \times 10^{11}$ years (106 ± 1 Byr) with ages obtained by the U-Pb (Pb-Pb) systems (for example, Lugmair

1974; Lugmair and Marti 1977; Lugmair, Scheinin, and Marti 1975), and because the ^{238}U and ^{235}U half-lives are more accurately known, the determination by Martins, Terranova, and Moreira Correa (1992) of $1.23 \pm 0.04 \times 10^{11}$ years (123 ± 4 Byr) should be viewed with caution, and indeed, that discrepant result most likely is an artifact. Begemann et al. (2001) noted that Martins, Terranova, and Moreira Correa (1992) reported in their experimental procedure the number of α -decays was registered on a "thin film of natural samarium oxide Sm_2O_3 with an overall uniform thickness of $(0.207 \pm 0.005) \text{ mg/cm}^2$." However, in their subsequent calculation of the decay constant, Martins, Terranova, and Moreira Correa (1992) apparently used the same thickness as for pure samarium element instead of correcting for oxygen. If this was a mistake as Begemann et al. (2001) claimed, then the published half-life value should have been multiplied by the weight ratio $\text{Sm}_2/\text{Sm}_2\text{O}_3 = 0.8624$, provided the Sm_2O_3 used was truly stoichiometric. This would have yielded a ^{147}Sm half-life value of 1.06×10^{11} years (106 Byr), which is in very good agreement with the previous five direct counting measurements (table 1). Begemann et al. (2001) reported that according to a private communication with one of the authors of the Martins, Terranova, and Moreira Correa (1992) paper, it is very likely that this explanation is correct, although it is not possible to give a definitive answer. For this and other reasons, Begemann et al. (2001) stated that they do not share the authors' opinion that their result "may be considered as the most accurate measurement of the half-life performed up to now" (that is, up to 1992). Nevertheless, Su et al.

(2010) performed parallel determinations using Sm metal and Sm_2O_3 (as described above) and obtained similar results of $1.06 \pm 0.01 \times 10^{11}$ years (106 ± 1 Byr) and $1.07 \pm 0.01 \times 10^{11}$ years (107 ± 1 Byr) respectively (table 1).

What then can be said about the Kinoshita, Yokoyama, and Nakanashi (2003) determination of $1.17 \pm 0.02 \times 10^{11}$ years (117 ± 2 Byr) (table 1)? Actually, it can be argued that their careful experimental approach using multiple repeated measurements with multiple ^{147}Sm sources using both an alpha spectrometer and a liquid scintillation spectrometer to measure the number of emitted α -particles per unit time, plus their detailed analysis of the counting efficiencies of the instrumental techniques they used, makes their determinations of the ^{147}Sm half-life more robust than any other determinations. Kossert et al. (2009) subsequently questioned the Kinoshita, Yokoyama, and Nakanashi (2003) determined ^{147}Sm half-life value because it did not agree with many other determined values. They also incorrectly asserted that Kinoshita, Yokoyama, and Nakanashi (2003) did not measure the isotopic ratio when their paper clearly reports that they did. These are not valid reasons to dismiss their determined ^{147}Sm half-life value. Worse still, rather than engage in discussion, Su et al. (2010) simply ignored the Kinoshita, Yokoyama, and Nakanashi (2003) determined ^{147}Sm half-life value. Yet the reality is that the Kinoshita, Yokoyama, and Nakanashi (2003) determined ^{147}Sm half-life value of 117 ± 2 Byr is in very good agreement with the four direct counting values of 114 ± 5 Byr of Karras and Nurmia (1960), 117 ± 5 Byr of Karras (1960), and 115 ± 5 Byr of Gupta and MacFarlane (1970) who all used ionization chambers for their determinations, and of 113 Byr of Graeffe and Nurmia (1961). However, it seems that the accepted ^{147}Sm half-life value of 106 Byr is determined by “majority vote” because it is supported by the seven direct counting determinations of Al-Bataina and Jänecke (1987), Donhoffer (1964), Gupta and MacFarlane (1970), Kossert et al. (2009), Su et al. (2010), Valli et al. (1965), and Wright, Steinberg, and Glendenin (1961) (table 1).

Kinoshita, Yokoyama, and Nakanashi (2003) were well aware that the ^{147}Sm half-life values reported from 1954 to 1970 showed some scatter between 104 Byr and 128 Byr, so they purposed to design their experimental procedures to thoroughly reevaluate the ^{147}Sm half-life by means of a developed alpha spectrometer with counting sources prepared in a manner different from those adopted in those earlier determination experiments. Two kinds of commercially available Sm_2O_3 reagents of 99.9% purity and two kinds of commercially available Sm standard solutions for atomic absorption

spectroscopy were prepared as four standard Sm solutions (with known concentrations in $\mu\text{g-Sm/g-solution}$). These were labelled as Sm-A, Sm-B, Sm-C, and Sm-D. Radioactive and non-radioactive impurities in these Sm solutions were measured by low-level gamma spectrometry, neutron activation, ICP-MS spectrometry, and thermal ionization mass spectrometry (TIMS), and it was confirmed that the solutions did not contain any detectable impurities, and that the isotopic composition was natural (that is, 15.0% ^{147}Sm).

Kinoshita, Yokoyama, and Nakanashi (2003) used calibrated solutions of the α -radioactive nuclides ^{210}Po , ^{238}U , and ^{241}Am as internal standards in preparation of the counting sources for alpha spectrometry. Known aliquots of each of the four Sm standard solutions were mixed well with known amounts of the calibrated ^{210}Po , ^{238}U , and ^{241}Am standard solutions to produce ten kinds of combinations. Each mixture was then evaporated on watch glasses and the amount of Sm on each watch glass was adjusted to $\sim 100 \mu\text{g}$, which means that there was approximately $15 \mu\text{g}$ of ^{147}Sm on each watch glass. Alpha spectrometry, using a silicon surface-barrier detector with an active area of 450mm^2 , was carried out for one to two weeks for each of the counting sources, and the α -disintegration rate of the known amounts of ^{147}Sm was determined by reference to the α -activities of the internal standards.

Measurements of the α -disintegration rates of the known amounts of ^{147}Sm were also carried out concurrently by Kinoshita, Yokoyama, and Nakanashi (2003) using a liquid scintillation spectrometer. However, only the ^{241}Am internal standard solution was added to each of the four Sm standard solutions. Each mixture was prepared for the measurements and then 10 ml of either the Amersham ACS II scintillation cocktail or the p-terphenyl+POPOP+toluene scintillation cocktail was added. A blank sample for background liquid scintillation measurement was also prepared in the same way. The liquid scintillation counting was continued for five hours for each of the counting sources. The same cocktail mixture for establishing the quench curve was used in determining the counting efficiency.

Kinoshita, Yokoyama, and Nakanashi (2003) then calculated a series of ^{147}Sm half-life values based on the ^{147}Sm α -activity of each of the prepared counting sources calibrated against the α -activity of each internal standard measured by the alpha spectrometer, and these are plotted in Fig. 3, along with results from several earlier determinations. They also calculated a series of ^{147}Sm half-life values measured with the liquid scintillation spectrometer, and these are plotted in Fig. 4. The attached error

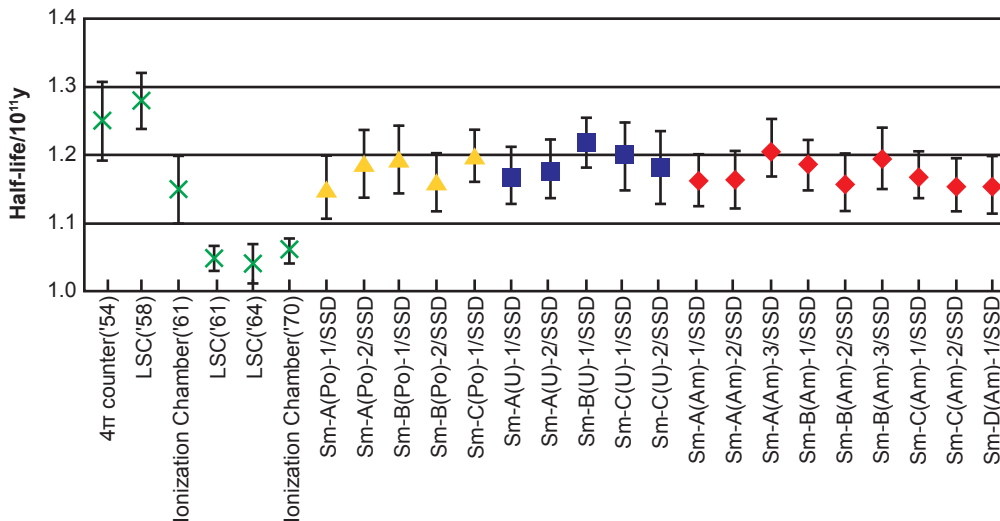


Fig. 3. The ¹⁴⁷Sm half-life values and their errors determined by alpha spectrometer using a silicon surface-barrier detector (after Kinoshita, Yokoyama, and Nakanashi [2003]). The values from earlier determinations are indicated by cross symbols, and values obtained by Kinoshita, Yokoyama, and Nakanashi (2003) are indicated by closed symbols. The prepared sources are referred to as the sample names with the original solution name, the reference standard, source number, and the measurement method.

for each plotted half-life value included the nominal systematic errors in the calibration of the α -emitting standard and in the measurement of the isotopic abundance of ¹⁴⁷Sm, in addition to the statistical error of 1 σ in α -counting. It is quite obvious from Figs. 3 and 4 that the ¹⁴⁷Sm half-life values obtained by Kinoshita, Yokoyama, and Nakanashi (2003) using an alpha spectrometer and a liquid scintillation spectrometer respectively agree very closely with each other within the error bars shown.

Kinoshita, Yokoyama, and Nakanashi (2003) were also careful to discuss the sources of error and their potential adverse effects on their ¹⁴⁷Sm half-life determination. They admitted that, in alpha spectrometry for a deposited counting source, self-

absorption of α -particles in the source is a serious problem. However, the energy loss and absorption of α -particles in the window-layer of the silicon surface-barrier detector (~50 nm) and in the vacuum chamber (<4 Pa) are negligible. As to the extent of absorption in their alpha spectrometry measurements, they argued that since the thickness of the residue of Sm and the α -emitting standard on the watch glass was less than 15 $\mu\text{g}/\text{cm}^2$, energy loss and absorption of α -particles from ¹⁴⁷Sm and the α -emitting standard of the counting source was expected to be negligible. On the other hand, while the counting efficiency of the liquid scintillation spectrometer is usually high (~100%), the spectrometer has the disadvantages of high background and poor energy-resolution.

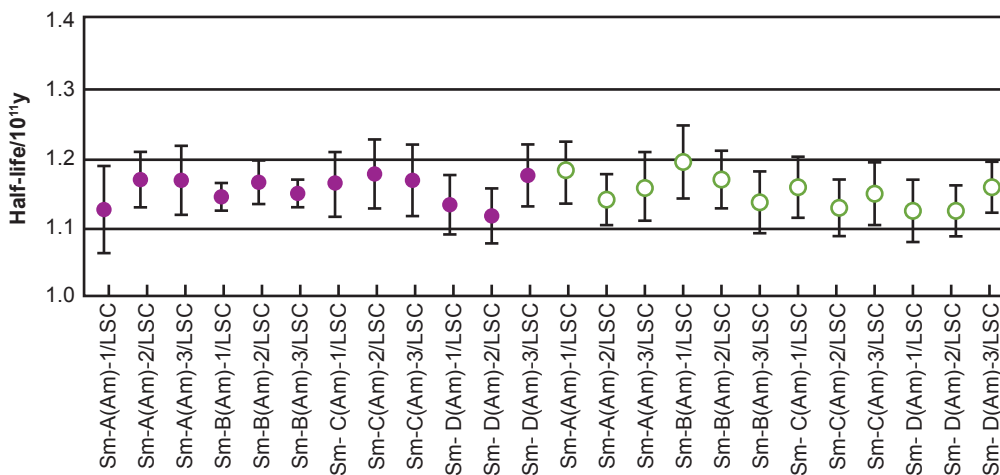


Fig. 4. The ¹⁴⁷Sm half-life values and their errors determined by liquid scintillation spectrometer (after Kinoshita, Yokoyama, and Nakanashi [2003]). The prepared sources are depicted as in Fig. 2. The Amersham ACS II scintillation cocktail is shown as closed symbols, and the p-terphenyl + POPOP + toluene scintillation cocktail is denoted with open symbols.

Hence, they concluded that in spite of the agreement between the mean ^{147}Sm half-life value of 115 ± 2 Byr measured with the liquid scintillation spectrometer and the mean ^{147}Sm half-life value of 117 ± 2 Byr measured with the surface-barrier detector, within the error, the former value was regarded as supporting data for the latter value because of these described disadvantages.

Kinoshita, Yokoyama, and Nakanashi (2003) determined that the arithmetic mean of the ^{147}Sm half-life values obtained in their experimental work for the 19 counting sources by alpha spectrometry (fig. 3) was 117 ± 2 Byr, the stated associated error being one standard deviation. This average ^{147}Sm half-life value was thus about 10% longer than the then, and still, currently adopted value of 106 ± 2 Byr. However, Kinoshita, Yokoyama, and Nakanashi (2003) noted that in the earlier determinations the half-life of ^{147}Sm was obtained by measuring the ^{147}Sm α -activity with a 4π gas-flow counter (Beard and Wiedenbeck 1954), liquid scintillation spectrometers (Beard and Kelly 1958; Donhoffer 1964; Wright, Steinberg, and Glendenin 1961), and ionization chambers (Gupta and MacFarlane 1970; MacFarlane and Kohman 1961) from sources with the number of ^{147}Sm atoms in them also measured. Because the ^{147}Sm half-life is then calculated conventionally from the obtained values of the ^{147}Sm α -activity and the number of ^{147}Sm atoms, the experimental errors in measuring those two values result in inaccurate determinations of the ^{147}Sm half-life. For example, impurities in the Sm reagent bring error into the number of ^{147}Sm atoms in the counting source, and uncertainty in the counting efficiency, self-absorption of the counting source and radioactive impurities in the Sm reagent bring error into the value of the ^{147}Sm α -activity. Kinoshita, Yokoyama, and Nakanashi (2003) thus suggested that the purity of the Sm reagent was not sufficient in the earlier experimental determinations. They also suspected that the corrections for counting efficiency and self-absorption were not appropriate in the earlier experimental determinations. High background and poor energy resolution of the liquid scintillation spectrometers used in the earlier experimental determinations might also have resulted in inaccurate ^{147}Sm half-life values. Since Kinoshita, Yokoyama, and Nakanashi (2003) were confident that all these sources of error were excluded from their experimental work, in contrast to the earlier experimental determinations, they concluded that their result of 117 ± 2 Byr for the ^{147}Sm half-life is reliable.

As already noted, Kossert et al. (2009) subsequently questioned the Kinoshita, Yokoyama, and Nakanashi (2003) result of 117 ± 2 Byr for the ^{147}Sm half-life. They were confident that they had accurately determined

the ^{147}Sm half-life as being 107 ± 0.9 Byr because they had expended great effort to evaluate the liquid scintillation counting efficiency to be 100%, because the number of ^{147}Sm atoms in the Sm reagent had been measured by means of ICP-OES (inductively coupled plasma optical emission spectrometry) using a reference standard from the National Institute of Standards and Technology, and because their result agreed well with most of the other measurement results and the recommended value. However, as has been already noted, a significant number of other measurement results also disagree with the recommended ^{147}Sm half-life value of 106 ± 2 Byr and are closer to the Kinoshita, Yokoyama, and Nakanashi (2003) result of 117 ± 2 Byr. Furthermore, while Kossert et al. (2009) asserted that Kinoshita, Yokoyama, and Nakanashi (2003) measured quite low activities in their experiments, they incorrectly accused Kinoshita, Yokoyama, and Nakanashi (2003) of not measuring the Sm isotopic ratio when they clearly stated that they did. Such an error would cast doubt on the Kossert et al. (2009) assessment of the experimental work of Kinoshita, Yokoyama, and Nakanashi (2003), especially as Kossert et al. (2009) admitted they did not have enough information on the Kinoshita, Yokoyama, and Nakanashi (2003) measurement details, in particular on the treatment of the background and tailing of the α -peaks in their measured spectra. But Kinoshita, Yokoyama, and Nakanashi (2003) did provide measurement details, as already described, and the α -peaks in their spectra measured by alpha spectrometer are very narrow, sharp, and distinct with a short tail and very low background, as illustrated in their report. So the reality is that Kossert et al. (2009) questioned the Kinoshita, Yokoyama, and Nakanashi (2003) result primarily because it didn't agree well with most of the other recent measurement results and the recommended value, even though Kinoshita, Yokoyama, and Nakanashi (2003) did 19 alpha spectrometer determinations using combinations of four standard Sm solutions and three internal standards (fig. 3) backed up by 24 liquid scintillation spectrometer determinations yielding essentially the same results (fig. 4).

On the other hand, Su et al. (2010) simply ignored the Kinoshita, Yokoyama, and Nakanashi (2003) determined ^{147}Sm half-life value of 117 ± 2 Byr after an introductory passing reference to it, and simply asserted that the "absolute" half-life they had determined, 106 ± 1 Byr (Sm metal) and 107 ± 1 Byr (Sm_2O_3), is consistent with the recommended value of 106 Byr. Yet Su et al. (2010) used a silicon surface-barrier detector coupled to an alpha spectrometer to similarly obtain their ^{147}Sm α -activity peaks, just as Kinoshita, Yokoyama, and Nakanashi

(2003) had done. And Su et al. (2010) tabulated the uncertainty components in both their ^{147}Sm α -activity measurements and their measurements of the number of ^{147}Sm atoms in their counting sources, which together summed to 1.1% of the ^{147}Sm half-life value, close to the figure of 0.9% uncertainty in the Kossert et al. (2009) determination, but considerably less than the ~2% obtained by Kinoshita, Yokoyama, and Nakanashi (2003). It would thus seem that the major reason for the difference between the Su et al. (2010) and Kinoshita, Yokoyama, and Nakanashi (2003) results may be due to the different Sm reagents used and the different procedures in preparing the counting sources.

Su et al. (2010) used ^{147}Sm -enriched metal and Sm_2O_3 powder obtained from Oak Ridge National Laboratory and did not analyze them for purity, although they did perform isotope dilution mass spectrometry (IDMS) to check the ^{147}Sm abundance, whereas Kinoshita, Yokoyama, and Nakanashi (2003) simply used commercially available Sm_2O_3 reagents and analyzed them for both impurities and the ^{147}Sm abundance. Then Su et al. (2010) used vacuum evaporation (Sm metal) and sputtering (Sm_2O_3) to deposit the Sm reagents onto glass substrates to a thickness of about $250\mu\text{g}/\text{cm}^2$ before checking the uniformity of ^{147}Sm distribution in these prepared counting sources by exposure to plastic emulsions. On the other hand, Kinoshita, Yokoyama, and Nakanashi (2003) simply evaporated the prepared mixtures of Sm standard solutions and internal standards onto glass substrates adjusted to $\sim 15\mu\text{g}$ of ^{147}Sm to ensure calculated amounts of α -particles would be measured from the ^{147}Sm and internal standards in these prepared counting sources. However, neither the choice of Sm reagents nor the procedures in preparing the counting sources seem to be significantly different between these two ^{147}Sm half-life determinations, even though Kinoshita, Yokoyama, and Nakanashi (2003) stated they prepared their counting sources in a manner different from earlier experiments. Apart from Su et al. (2010) not analyzing their chosen Sm reagents for purity, which would not be expected to be significant given the source of those reagents, the only major difference appears to have been the thicknesses of the counting sources and therefore the numbers of ^{147}Sm atoms in them, resulting in only ~ 150 counts in the central channel of the α -peaks obtained by Kinoshita, Yokoyama, and Nakanashi (2003), the quite low activities in their experiments referred to by Kossert et al. (2009), compared to the 500–600 counts in the central channel of the α -peaks obtained by Su et al. (2010). Yet in spite of the lower α -activity, it could be argued that the thinner counting source should have yielded the more accurate determination

of the ^{147}Sm half-life value, because even though the thicker counting source would contain more ^{147}Sm atoms and therefore emit more α -particles, the thicker the counting source the more self-absorption of α -particles there could be within the counting source thus significantly reducing the determined ^{147}Sm half-life value.

If indeed the thicknesses of the counting sources and thus the self-absorption of α -particles in them have resulted in the experimentally determined values of the ^{147}Sm half-life being up to 10% or more different, then this has profound significance on the determinations of the Sm-Nd radioisotope ages of rocks, minerals, and meteorites. Without certainty as to whether the present ^{147}Sm half-life (decay rate) has been accurately determined experimentally by direct counting, and the assumption of constant radioisotope decay rates at the currently determined values, the Sm-Nd radioisotope ages calculated by uniformitarians cannot be accurately known, no matter how accurate are the measured $^{147}\text{Sm}/^{144}\text{Nd}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios in rocks, minerals, and meteorites.

In any case, the adopted ^{147}Sm half-life value of $106\pm 0.8\text{Byr}$ had essentially been already settled on the basis of the geological comparisons done on two meteorites by Lugmair, Scheinin, and Marti (1975) and Lugmair and Marti (1977) who adjusted these meteorites' Sm-Nd ages to agree with their Pb-Pb ages (Begemann et al. 2001; Dickin 2005, 70–71). Lugmair (1974) compared the Sm-Nd isochron age he obtained for the Juvinas eucrite meteorite with the similar Rb-Sr isochron age obtained on the same meteorite by Allègre, Birck, and Fourcade (1973). However, it was Lugmair, Scheinin, and Marti (1975) whom Dickin (2005, 70–71) credited for the Sm-Nd isochron age of $4.56\pm 0.08\text{Byr}$ for Juvinas that is in agreement with the Pb-Pb isochron age for the solar system and thus confirming the adopted value of 106Byr for the ^{147}Sm half-life (Dickin 2005, 115–118; Patterson 1956). This though begs the question—how do they know that these so-called isochrons are not mixing lines which have no time significance? It should also be noted that Dickin (2005, 71, fig. 4.1 caption) wrote that the “Nd isotope ratios are affected by the choice of normalizing factor for mass fractionation,” a reminder that the agreement of this Sm-Nd isochron age with the Pb-Pb isochron age was only achieved by making choices of suitable factors. Subsequently, Lugmair and Marti (1977) obtained a Sm-Nd isochron age of $4.55\pm 0.04\text{Byr}$ for the Angra dos Reis (ADOR) angrite meteorite based on analyses of phosphate and pyroxene mineral separates, which they made sure agreed with the Pb-Pb model age of $4.555\pm 0.005\text{Byr}$ for ADOR obtained by Tatsumoto, Knight, and Allègre (1973). Lugmair and Galer

(1992) later refined that Pb-Pb model age for ADOR to 4.55780 ± 0.00042 Byr based on analyzing a pyroxene mineral separate, which they then used to suggest what the ^{147}Sm half-life should be in order to make the Sm-Nd isochron age for this meteorite agree exactly with this Pb-Pb model age.

Lugmair and Marti (1977) and Lugmair and Galer (1992) justified their geological comparisons to adjust the ^{147}Sm half-life value so that the Sm-Nd isochron ages of the Juvinas and Angra dos Reis meteorites agreed with their Pb-Pb isochron and model ages on the basis that these meteorites' Pb-Pb ages were more precise because the decay constants of the parent ^{238}U and ^{235}U "are known more precisely." And Lugmair and Marti (1978) added to the adoption of that ^{147}Sm half-life value of 106 Byr by choosing the "most precise" results from only four of the earlier direct counting experiments (see table 1) that would give the desired weighted average value for the ^{147}Sm half-life of 106 ± 0.8 Byr "that has been adopted by all geo- and cosmochronologists since that time" (Begemann et al. 2001). This was also the basis for Begemann et al. (2001) seeking to "correct" the Martins, Terranova, and Moreira Correa (1992) determined higher ^{147}Sm half-life value to bring it into agreement with this adopted ^{147}Sm half-life value calibrated against the U-Pb radioisotope system, and for Kossert et al. (2009) and Su et al. (2010) dismissing and ignoring respectively the more thoroughly determined but higher ^{147}Sm half-life value of Kinoshita, Yokoyama, and Nakanashi (2003).

However, the U-Pb "gold standard" dating method has come under much scrutiny in the two last decades. Ludwig (2000) has demonstrated that, although almost universally ignored, the effect of the errors in the U decay constants on U-Pb concordia-Pb-Pb intercept ages are significant, being a 4.5 Myr error for a 500 Myr age, which amounts to almost a 1% error. The U-Pb method also depends on the crucial $^{238}\text{U}/^{235}\text{U}$ ratio, but discrepancies and variations have been found recently between the $^{238}\text{U}/^{235}\text{U}$ ratio in U-bearing terrestrial minerals and rocks and the $^{238}\text{U}/^{235}\text{U}$ ratio in meteorites (Brenneka and Wadhwa 2012; Hiess et al 2012). Much earlier, Apt et al. (1978) had reported that the $^{235}\text{U}/^{238}\text{U}$ ratio in uranium ores in Canada, Brazil, Zaire, and Australia varied from 0.7107 to 0.7144 when the recognized value is 0.72. Such variations in uranium ores have been further documented by Bopp et al. (2009). These discrepancies and variations remain unexplained, especially in the context of the ^{238}U and ^{235}U decay constants and half-lives. Furthermore, the fact that there are these variations in the crucial $^{238}\text{U}/^{235}\text{U}$ ratio in terrestrial minerals and rocks on which the U-Pb dating "gold standard" depends, which has been used to recalibrate Sm-Nd isochron ages to determine the

^{147}Sm half-life and decay constant, only underscores that these radioisotope methods cannot provide the absolute invariable "dates" they are so confidently proclaimed to provide.

In any case, there is the additional assumption in all the radioisotope dating methods of having to know the original concentrations of the daughter and index isotopes which is very significant in the U-Pb method because the original concentrations are assumed not to be zero, in contrast to the K-Ar method. Yet there must be great uncertainty as to what those initial values were in the unobserved past, despite the isochron and concordia techniques attempting to negate the necessity for knowing those initial values, and despite the assumption ever since Patterson (1956) and Tatsumoto, Knight, and Allègre (1973) that the Pb isotopic composition of the troilite (FeS) in the Canyon Diablo iron meteorite represents the initial "primordial Pb" of the earth and the solar system (Dickin 2005; Faure and Mensing 2005). Thus the U-Pb method should not be used as a standard to determine other parent radioisotope half-lives and decay constants.

Indeed, it would remain prudent to be very careful with these geological comparison methods for two other reasons. First, there are significant flaws in the basic assumptions on which all the radioisotope dating methods depend, not least being the assumption that the decay rates of the parent radioisotopes have always been constant in the past at today's measured decay rates. Second, the U-Pb method relies primarily on α -decay, as does the Sm-Nd method. Yet both Austin (2005) and Snelling (2005) have reported that the parent U and Sm α -decaying radioisotopes seem to yield systematically different U-Pb and Sm-Nd ages for some earth rocks using the same samples with essentially the same methodology. Additionally, they suggested the pattern of differences was potentially related to the parent radioisotopes' atomic weights and half-lives, which could be indicative of parent radioisotopes' decay rates having not been constant in the past but instead were substantially faster. Furthermore, these different radioisotope ages yielded by the same earth rocks are often widely divergent, even up 100–200% different, which is such a huge divergence that it renders these dating methods highly suspect, even if the differences in the determinations of the half-lives of the parent radioisotopes seem miniscule and therefore trivial by comparison. However, it was considered prudent to still document here these seemingly miniscule differences in half-life values, because they may be indicative of other underlying factors at work (as already discussed briefly), and they can still lead to very significant discrepancies in the derived

radioisotope ages that might otherwise appear to be acceptably accurate to uniformitarians.

Nevertheless, the age comparisons on meteorites used the U-Pb method back in the 1970s to settle, apparently beyond any subsequent dispute, the determination of the ^{147}Sm decay half-life at $106 \pm 0.8 \text{ Byr}$, which is within the range determined by many of the physical direct counting experiments by several techniques (see table 1 and fig. 2). Yet the robustness of the 10% higher $117 \pm 2 \text{ Byr}$ ^{147}Sm half-life value determined by Kinoshita, Yokoyama, and Nakanashi (2003) might even indicate that the other higher ^{147}Sm half-life values determined in some earlier experiments (see table 1 and fig. 2) should not be simply dismissed as due to poorer experimental methodology or equipment. In any case, Sm-Nd age calculations are now ultimately calibrated against the U-Pb method, and thus the ^{147}Sm half-life value of $106 \pm 0.8 \text{ Byr}$ has been adopted. However, this U-Pb “gold standard” depends on whether the U decay constants are accurately and precisely known, and on the crucial $^{238}\text{U}/^{235}\text{U}$ ratio. Yet discrepancies and variations have been found between the $^{238}\text{U}/^{235}\text{U}$ ratio in U-bearing terrestrial minerals and rocks and the $^{238}\text{U}/^{235}\text{U}$ ratio in meteorites which remain unexplained. This only serves to highlight that if the Sm-Nd dating method has been calibrated against the U-Pb “gold standard” with its own uncertainties, then the claimed accurately-determined ^{147}Sm decay rate cannot be absolute, especially given the evidence in some earth rocks of past higher radioisotope decay rates and the evidence that some direct counting experiments yielded 10% or more higher ^{147}Sm half-life values. Yet even though it is to be expected these half-life measurements vary by 10% or so because of the difficulties in measuring such a long half-life, the resultant calculated radioisotope ages end up being an order of magnitude or more different from one another, which is far too “inaccurate” in providing the absolute ages required by uniformitarians. Thus without an accurately known ^{147}Sm decay half-life, accurate Sm-Nd radioisotope ages cannot be accurately determined. Therefore, Sm-Nd dating cannot be used to reject the young-earth creationist timescale, especially as current radioisotope dating methodologies are at best hypotheses based on extrapolating current measurements and observations back into an assumed deep time history for the cosmos.

Conclusions

There have been numerous attempts to determine the ^{147}Sm decay half-life in the last 80 years by two primary techniques used in direct physical counting experiments—ionization chambers and liquid scintillation counters, and by radioisotope age comparisons using two meteorites. The

determinations since 1960 have converged with close agreement on the ^{147}Sm half-life value of $106 \pm 0.8 \text{ Byr}$, which has since the 1970s been adopted for standard use by the uniformitarian geological community. This adopted ^{147}Sm half-life value is the weighted average of four determinations by direct counting experiments in the period 1961–1970, confirmed by geological comparisons in the 1970s in which the ^{147}Sm half-life value was adjusted in order to recalibrate (or force, essentially by circular reasoning) the Sm-Nd isochron ages of two meteorites to be the same as their Pb-Pb isochron and model ages.

However, even though this ^{147}Sm half-life value of $106 \pm 0.8 \text{ Byr}$ has been universally adopted by the geochronology and cosmochronology community since Lugmair and Marti (1978) proposed it (Begemann et al. 2001), the more recent direct counting experiments by Kinoshita, Yokoyama, and Nakanashi (2003) determined the ^{147}Sm half-life value was 10% or more longer at $117 \pm 2 \text{ Byr}$. They achieved this by using four standard Sm solutions with internal α -radioactive standards in 19 alpha spectrometer and 24 ionization chamber determinations, making it the most thorough and comprehensive effort to the determine the ^{147}Sm half-life. It would appear that the thinner counting sources they used, while resulting in low α -activities being measured, greatly reduced the counting uncertainty due to self-absorption of the emitted α -particles. So in spite of being rejected or ignored, this $117 \pm 2 \text{ Byr}$ value for the ^{147}Sm half-life, which agrees with some earlier determinations, may well be highly significant and more reliable than the adopted value.

Yet even though there is close agreement between many determined values, the ^{147}Sm half-life value obtained by recalibrating the Sm-Nd isochron ages for two meteorites with their Pb-Pb isochron and model ages has been given preference over the values obtained by direct counting experiments which directly measure ^{147}Sm decay, and so are independent of all the assumptions involved with the radioisotope dating methods. Indeed, model dependent results should not take precedence over the direct experimental evidence.

Since the age comparisons on two meteorites used the U-Pb method to determine the ^{147}Sm decay half-life, all Sm-Nd age calculations are thus ultimately calibrated against the U-Pb method. However, this U-Pb “gold standard” depends on having precisely determined ^{238}U and ^{235}U decay constants, as well as on the crucial $^{238}\text{U}/^{235}\text{U}$ ratio being known and constant. Yet there are still uncertainties in the measured U decay constants, and discrepancies and variations have been found between the $^{238}\text{U}/^{235}\text{U}$ ratio in U-bearing terrestrial minerals and rocks, and the $^{238}\text{U}/^{235}\text{U}$ ratio in meteorites. These discrepancies and

variations remain unexplained. This only serves to highlight that if the Sm-Nd dating method has been calibrated against the U-Pb “gold standard” with its own uncertainties, then it cannot be absolute.

Furthermore, there is evidence that nuclear decay rates have not been constant in the past. Thus without an accurately known ^{147}Sm decay half-life, accurate Sm-Nd radioisotope ages cannot be accurately determined. Therefore, Sm-Nd dating cannot be used to reject the young-earth creationist timescale, especially as current radioisotope dating methodologies are at best hypotheses based on extrapolating current measurements and observations back into an assumed deep time history for the cosmos.

Acknowledgments

The invaluable help of my research assistant Lee Anderson Jr. in compiling and plotting the data in the color-coded age diagrams is acknowledged. The three reviewers are also acknowledged for their helpful comments and input, though the final content of this paper is solely my responsibility. Our production assistant Laurel Hemmings is also especially thanked for her painstaking work in preparing this paper for publication.

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