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THE USE OF Sm-146 IN AGE DETERMINATIONS

by

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Abstract

On the basis of the recently reported discovery of Sm-146 in natural Sm, it is estimated that its half-life must be in the vicinity of $3 \cdot 10^8$ years. From nuclear abundances it is concluded that the initial isotopic abundance of Sm-146 was between 1 and 10%. Important information about the age of the universe can be obtained once its half-life becomes accurately known.

1. *Introduction*

The isotope samarium-146 was discovered by Dunlavey and Seaborg [2], who bombarded neodymium metal with alpha particles and detected a 2.55-MeV alpha activity, which was ascribed to Sm-146. Its half-life was estimated by the same investigators to be $5 \cdot 10^7$ years.

The presence of Sm-146 in natural Sm was recently discovered by Vorobev and collaborators [9] by measuring the alpha spectrum of natural Sm with an ionization chamber. In addition to the well known 2.20-MeV alpha-particles of Sm-147 [3], a weak activity was observed at 2.55 MeV and interpreted as being due to Sm-146. The presence of this nuclide in natural Sm has thus far escaped detection by mass spectrometric means, the upper limit for its abundance given by Collins and collaborators [1] being $8 \cdot 10^{-5}$%.

The possible geochronological importance of Sm-146 becomes clear when it is noted that it is by far the shortest-lived natural radionuclide that is not being continuously produced in nature. The next in this respect is U-235, with a half-life of $7 \cdot 10^8$ years, and Sm-146 may thus yield important information about the age of the earth and of the universe if the necessary data can be found.

2. *The Half-Life of Sm-146*

In the case of nuclear species with half-lives too long for direct measurement the specific activity s, i.e. the number of disintegrations taking place per gram of substance per unit time (in this case a year), is measured. The half-life $t_{1/2}$ is then obtained from the equation

$$t_{1/2} = \frac{0.693}{\lambda} = \frac{0.693 \cdot N}{M \cdot s}$$

where N is Avogadro's number and M the mass number of the nuclide.

The only result thus far published concerning Sm-146 is that of DUNLAVEY and SEABORG (loc.cit.), who estimated the amount of Sm-146 present on the basis of the probable yield on bombardment. Their result, $5 \cdot 10^7$ years, is thus a very approximate one, and it is clearly in disagreement with the findings of VOROBEV and collaborators. From the data published by the latter investigators it can be estimated that the present ratio of the activities of Sm-146 and Sm-147 is $5 \cdot 10^{-4}$. Thus

$$\frac{\lambda_{146}}{\lambda_{147}} \cdot \frac{a_{146}}{a_{147}} = 5 \cdot 10^{-4}$$

where the λ's denote the decay constants and the a's the present abundances of the two nuclides. Now $\lambda_{147} = 6.0 \cdot 10^{-12}$ per year [3,4] and $a_{147} = 15.09\%$ [1], and we obtain

$$\lambda_{146} \cdot a_{146} = 4.5 \cdot 10^{-14} \frac{\%}{\text{year}} .$$

We can now calculate the original abundance of Sm-146 as a function of its assumed half-life if the time elapsed since the synthesis of the elements is known. The results are given in Fig. 1 for the cases that the age of the universe is assumed to be 4, 5, or $6 \cdot 10^9$ years. It is immediately evident that the half-life of Sm-146 must be appreciably longer than $5 \cdot 10^7$ years, and if it is assumed that the original abundance of Sm-146 was

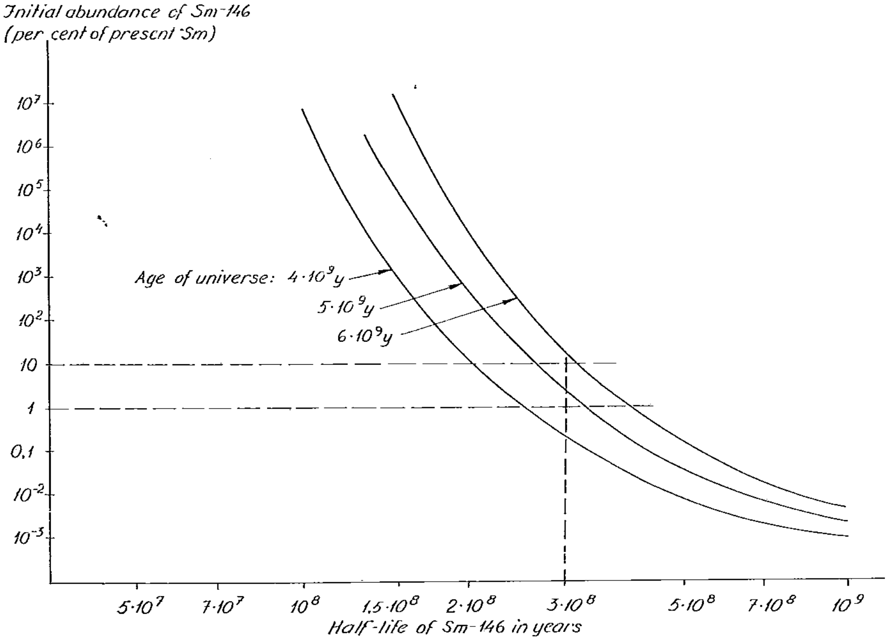


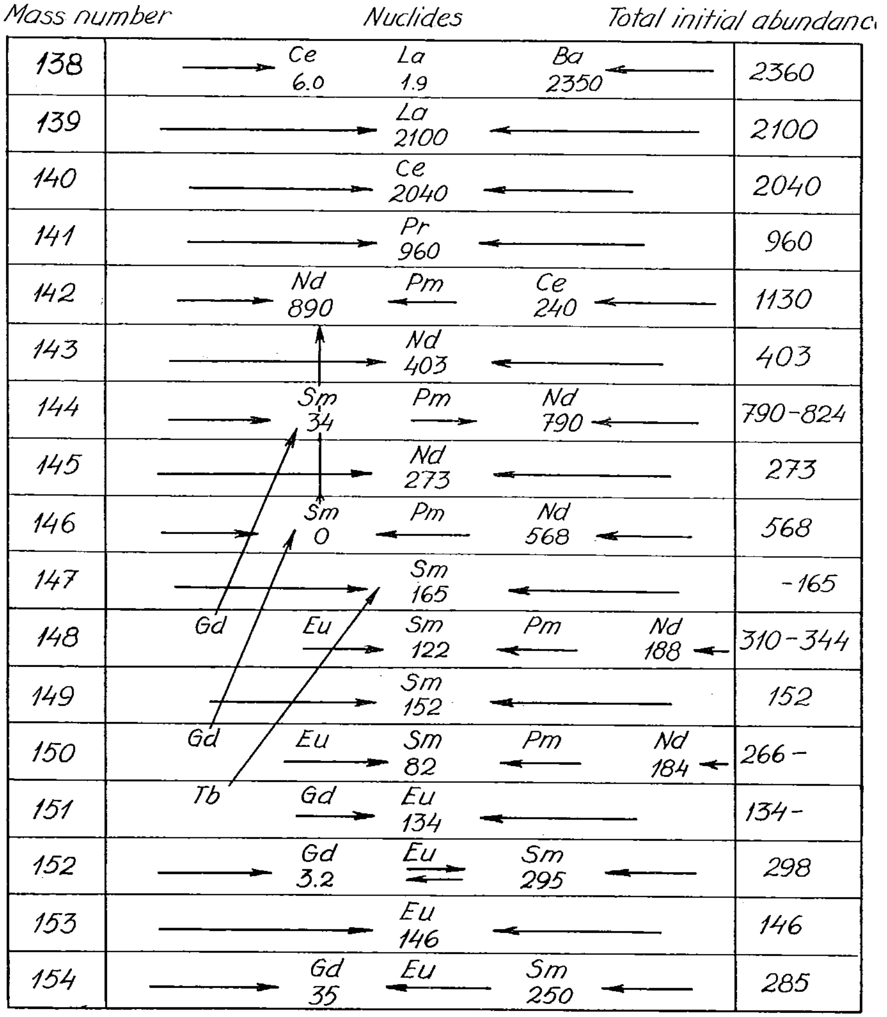
Fig. 1. Initial isotopic abundance of Sm-146 in per cent of the total present Sm as a function of its assumed half-life, calculated for different ages of the universe.

between 1 and 10% (see below), the most probable value of its half-life is found to be $3 \cdot 10^8$ years. As it is probable that the half-life can be independently determined in the near future, the question of estimating its initial abundance becomes quite important.

3. *The Initial Abundance of Sm-146*

Since Sm-146 decays to Nd-142, an absolute upper limit for its initial abundance is readily obtained by assuming that all Nd-142 has been formed from Sm-146. The abundance of Nd-142 in natural Nd is 27.1% [8], and since the amount of Nd present in the universe is estimated to be three times that of Sm (7), the upper limit for Sm-146 becomes equal to 100 in the units employed in Fig. 1.

A better estimate can be obtained by studying the relative abundances of different nuclides in the region in question. It is generally assumed that the synthesis of the medium-weight nuclides took place by some sort



Mass number	Nuclides	Total initial abundanc
138	→ Ce 6.0 La 1.9 Ba 2350 ←	2360
139	→ La 2100 ←	2100
140	→ Ce 2040 ←	2040
141	→ Pr 960 ←	960
142	→ Nd 890 Pm ← Ce 240 ←	1130
143	→ Nd 403 ←	403
144	→ Sm 34 Pm → Nd 790 ←	790-824
145	→ Nd 273 ←	273
146	→ Sm 0 Pm ← Nd 568 ←	568
147	→ Sm 165 ←	-165
148	Gd Eu → Sm 122 Pm ← Nd 188 ←	310-344
149	→ Sm 152 ←	152
150	Gd Eu → Sm 82 Pm ← Nd 184 ←	266 -
151	Tb Gd → Eu 134 ←	134-
152	→ Gd 3.2 Eu ⇄ Sm 295 ←	298
153	→ Eu 146 ←	146
154	→ Gd 35 Eu ← Sm 250 ←	285

Fig. 2. Beta and alpha decay chains in the rare earth region, with present relative abundances of the nuclides found in nature.

of catastrophic process in a very short time, and it is natural to assume that both stable and radioactive nuclei were formed in amounts that bear a statistical relation to their stability under the conditions during their formation.

The initial pattern of abundances produced in the primordial act subsequently was modified by beta decay of all radioactive chains leading

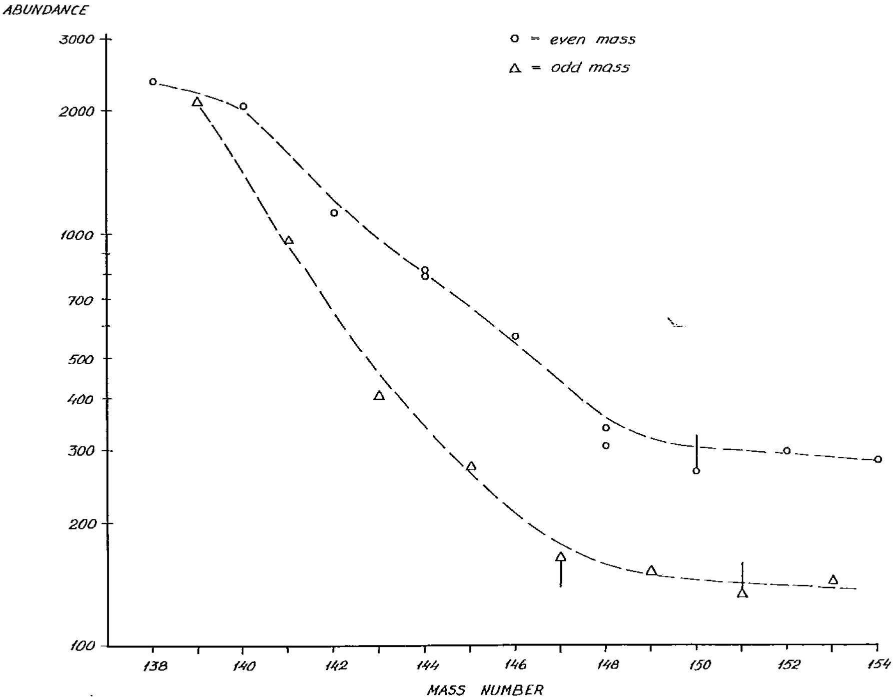


Fig. 3. Relative abundances of different mass numbers in nature.

to stable or very long-lived products. In addition, further modification of the abundance pattern is caused by alpha emission which in certain cases connects collateral chains. This is illustrated in Fig. 2, which also contains relative abundance values for each nuclide, obtained by multiplying its percentage abundance by the abundance of the element in question in the universe (7). The complications caused by alpha-decay are marked with vertical arrows, and it is seen that Gd-150 decays into Sm-146, which in turn decays into Nd-142.

The combined present abundance of each mass number is plotted in Fig. 3. These data are based on zero abundance of Sm-146, and it is seen that the point for mass 146 falls slightly above the curve even without the Sm-146. The point for the decay product 142 does not show any excess that could be attributed to the decay of Sm-146, and the formation of this nuclide in substantial amounts by the alpha decay of Gd-150 also seems quite unlikely because there is very little defect in the amount of mass 150 present. On this basis it is concluded that the initial abundance

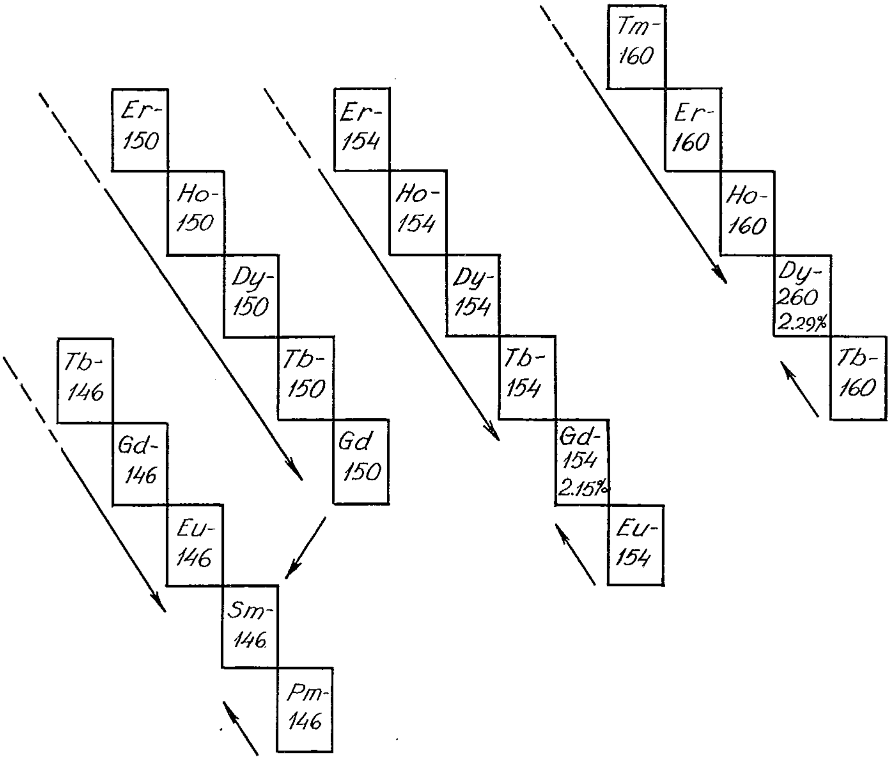


Fig. 4. Decay chains leading to Sm-146, Gd-154, and Dy-160.

of Sm-146 probably was less than 100 in the units of figs. 2 and 3, corresponding to an initial isotopic abundance of 10%.

On the other hand, it seems unlikely that the initial abundance of Sm-146 was much less than that of the corresponding Gd isotope (2.15%) or Dy isotope (2.29%). Like Sm-146, both are products of the decays of all nuclides in a region of the nuclide chart that includes one position on the neutron-excess side and the whole isobar line on the neutrondeficient side (see Fig. 4). For Sm-146 the latter part contains, in addition, the collateral chain terminating at Gd-150, and it is thus concluded that the initial abundance of Sm-146 probably was greater than 1%.

4. *The Use of Sm-146 in Geological Dating*

For the purposes of this discussion we assume that the half-life of Sm-146 is $3 \cdot 10^8$ years and that the elements originated $5 \cdot 10^9$ years ago.

These quantities fix the initial isotopic abundance of Sm-146 as 3% (see Fig. 1), while its present isotopic abundance would be $3 \cdot 10^{-5}\%$, in agreement with the upper limit ($8 \cdot 10^{-5}\%$) given by COLLINS and collaborators. This means that the abundance of Sm-146 $3 \cdot 10^9$ years ago was about 0.01%, and that a Sm mineral of this age should contain a corresponding excess of the decay product Nd-142.

Most such minerals contain appreciably more Nd than Sm, but the selective assemblage-types often contain about three times as much Sm as Nd [6], and in such a mineral an excess of 0.03% in the isotopic abundance of Nd-142 would result from an age of $3 \cdot 10^9$ years. Since the abundance of Nd-143 should simultaneously exhibit an excess of about 1% due to the decay of Sm-147, it is seen that under the assumptions made, Sm-146 would be useful in case of extremely old samples only.

Among the pleochroic rings or halos caused by the action of alpha particles emitted from small radioactive inclusions in certain minerals, the so-called L rings correspond to an alpha-particle range of 1.18 cm in air and are believed to be caused by Sm-147 [5].

Because the activity caused by Sm-146 was equal to that caused by Sm-147 about $3.3 \cdot 10^9$ years ago, one would predict that somewhat larger rings corresponding to the higher alpha range of Sm-146 (1.35 cm) should be found associated with the L-rings in sufficiently old minerals.

It may thus be concluded that Sm-146 is likely to become important in geological dating in exceptional cases only. On the other hand, it may yield important information about the age of the universe if its half-life can be accurately determined.

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