

this La_2O_3 , two samples were weighed out and then sealed in plastic holders for use in our studies of ^{138}La decay. The La sources were counted using a large volume Ge(Li) detector, and from the measured γ -ray yields, the β -decay and electron capture (EC) decay partial half lives were determined. Subsequent tests on separate samples of this same La_2O_3 showed that when this material is exposed to air for a period of several days, 10–15% weight increases are observed. These weight increases are probably due to the absorption of H_2O and CO_2 as suggested by Tanaka and Masuda¹. However, this process occurs only in material left exposed to air, and thus does not affect the results of our half life measurements.

Our results for the ^{138}La β -decay, EC decay and total half lives are $3.19 \pm 0.22 \times 10^{11}$ yr, $1.58 \pm 0.02 \times 10^{11}$ yr, and $1.06 \pm 0.03 \times 10^{11}$ yr, respectively². These values are in excellent agreement with those recently obtained by Sato and Hirose³. As we have previously discussed², the relatively poor energy resolution of NaI detectors and the nearly unavoidable presence of radioactive impurities in La compounds may have led early investigators to obtain incorrect values for the ^{138}La half lives. Thus one probably should not include the results of such measurements in estimates of the ^{138}La half lives. The only experiments in which Ge(Li) detectors were used and in which attention was paid to the problem of volatile impurities in La_2O_3 samples were ours² and that of Sato and Hirose³. Unfortunately, Sato and Hirose quote only statistical uncertainties in their results³. However, the techniques used in their experiments were very similar to ours. Thus if we assume that their total uncertainty (statistical + systematic) is equal to ours, then we can combine the results of these two experiments. The mean of these two measurements yields a ^{138}La β -decay partial half life of $3.10 \pm 0.15 \times 10^{11}$ yr. Thus the discrepancy between the ages inferred from the $^{138}\text{La}/^{138}\text{Ce}$ and $^{147}\text{Sm}/^{143}\text{Nd}$ chronometers is slightly larger than was originally found by Tanaka and Masuda¹. The origin of this discrepancy remains an open question.

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TANAKA AND MASUDA REPLY—
Norman and Nelson have carefully redetermined the half life of ^{138}La using

a La_2O_3 source¹. They have presented 3.10×10^{11} yr for the partial half life of ^{138}La β -decay by combining their result¹ with that of Sato and Hirose², but still there appears to be some discrepancy between the ages inferred from the ^{138}La – ^{138}Ce and ^{147}Sm – ^{143}Nd chronometers³.

We have re-examined the Ce isotopic composition of the same sample powders as studied previously³, because our technique on the isotopic measurement has recently been improved. New results yield an age of $2,350 \pm 380$ (2σ) Myr based on the partial decay constant $2.58 \times 10^{-12} \text{ yr}^{-1}$ (half life of 2.69×10^{11} yr). Assuming instead a 13% longer partial half life of 3.10×10^{11} yr ($\lambda\beta^{138}\text{La} = 2.24 \times 10^{-12} \text{ yr}^{-1}$) advanced here by Norman and Nelson, the age comes out to be $2,710 \pm 430$ (2σ) Myr, which is even more discrepant compared with the ^{147}Sm – ^{143}Nd age ($2,050 \pm 90$ (2σ) Myr) and ^{87}Rb – ^{87}Sr age ($2,050 \pm 43$ (2σ) Myr)³. This implies that the inclination of our isochron line obtained for La–Ce systematics is too steep in view of the half life estimated from radioactivity.

If we suppose, on the other hand, that the discrepancy between ^{138}La – ^{138}Ce and ^{147}Sm – ^{143}Nd ages is real, a conceivable explanation is that this discrepancy was caused by alteration effects on the sample examined here. Lanthanum is considered among rare earth elements to be a relatively very mobile element on weathering or alteration. Sometimes the alteration brings about the increase in abundance of light rare earth elements in mid-Atlantic ridge basalts⁴. By addition of the same amounts of La to plagioclase and pyroxene, the $^{138}\text{La}/^{142}\text{Ce}$ ratio of pyroxene increases by a greater factor than that of plagioclase, because the La abundance of pyroxene (0.6–1.0 p.p.m.) is smaller than that of plagioclase (2.3–2.4 p.p.m.)³. The $^{138}\text{La}/^{142}\text{Ce}$ ratio of bulk sample will increase moderately and then the La–Ce isochron will become steeper, giving an apparently older La–Ce age. However, the La abundance is not likely to have been modified, because ^{87}Rb – ^{87}Sr and ^{147}Sm – ^{143}Nd ages agree despite the fact that Rb and Sr are considered to be more mobile elements than La on weathering or alteration.

There are several kinds of uncertainties which may have effects on the absolute age under consideration: (1) isotopic abundance of ^{138}La and ^{138}Ce ; (2) elemental abundances of La and Ce; (3) decay constant of ^{138}La . Uncertainties of isotopic abundance of ^{138}Ce and of elemental abundances of La and Ce were taken into consideration for age calculation³. Uncertainty of ^{138}La abundance was not considered. This uncertainty is estimated at $\sim 1.3\%$ (ref. 5) and it has an effect both on decay constant and on $^{138}\text{La}/^{142}\text{Ce}$ ratio in age calculation. However, uncertainties of the both terms are mathematically cancelled in the course of the age calculation.

Uncertainties in the decay constant should be evaluated from various sides. However, uncertainties for the most of the decay constants obtained previously (see Table 1 of ref. 3) have been estimated only from counting statistics. For the absolute evaluation and comparison between individual radioisotope clocks, all uncertainties on the procedures must be taken into account. For the accurate quantitative measurement of 789-keV γ ray, especially, which is an emission from excited ^{138}Ce , careful corrections such as detector efficiencies and relative attenuation by self-absorption compared with the standard γ -ray (for example, 1,461-keV γ -ray from ^{40}K) are required. From this standpoint, the 14% (2σ) uncertainty in γ -decay constant of ^{138}La by Norman and Nelson¹ seems to be the only reliable evaluation. If 10% (2σ) uncertainty estimated here by Norman and Nelson) uncertainty in the decay constant is taken into account for the age calculation, the resultant La–Ce age of the Bushveld gabbro falls in the range agreeable to the Sm–Nd age within experimental uncertainty. In this sense, we would again like to emphasize that it is essential to establish a precise and reliable value for the decay constant of ^{138}La . La–Ce geochronological studies which compare the Sm–Nd clock for some material older than the Bushveld gabbro such as meteorite will give that refinement.

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