

Origin of life from apatite dating?

Mojzsis *et al.*^{1,2} reported the carbon-isotope composition of carbonaceous inclusions in grains of apatite from sediment sequences of Akilia island, southwest Greenland, that are more than 3,850 million years (Myr) old. The $\delta^{13}\text{C}$ values measured by Mojzsis *et al.*¹ led them to conclude that these carbonaceous materials are evidence of early life 3,850 Myr ago. But if other isotopes (U–Pb and Pb–Pb) are used, the apatites are estimated at just $1,504 \pm 336$ (2σ) and $1,459 \pm 160$ (2σ) Myr old. This value is consistent with measurements of 1,600–1,700 Myr for Rb–Sr mineral isochrons on biotites³ and 1,670 Myr for K–Ar muscovite⁴ from Amitsoq gneiss in the region. We conclude that, about 1,500 Myr ago, these apatites in Akilia island experienced a metamorphic event of about 600 °C (estimated by the closure temperature of the U–Pb system^{5,6}).

Mojzsis *et al.*¹ measured the carbon-isotope composition of graphite inclusions in grains of apatite in banded-iron formations (BIFs) from Akilia island. Their observed $\delta^{13}\text{C}$ values ranged from –20 per mil (‰) to –50‰, using a PDB standard, indicating a biogenic origin. The BIFs are older than 3,850 Myr (ref. 2), but the age of the apatite housing the graphite material was not determined. Here we present measurements of the U–Pb age of apatites from closely related samples.

We cast the sample chip of the Akilia BIFs (approximately 1×1 cm) into epoxy-resin disks with several grains of standard apatite and polished them until they were exposed through their mid-sections. The sample apatites are about 20–30 μm in size and show similar texture to those reported by Mojzsis *et al.*¹. We focused a primary beam of about 2.5 nA O_2^- to sputter an area of apatites 20 μm in diameter, and extracted the positive secondary ions using 10 kV. We found no isobaric interferences in the mass range over ^{204}Pb and ^{208}Pb at a mass resolution of 5,800. We obtained the $^{238}\text{U}/^{206}\text{Pb}$ ratios from the observed $^{238}\text{U}^+/^{206}\text{Pb}^+$ ratios by calibration using an empirical quadratic relationship between $^{206}\text{Pb}^+/^{238}\text{U}^+$ and $^{238}\text{U}^{16}\text{O}^+/^{238}\text{U}^+$ ratios of standard. The experimental details of the apatite U–Pb analysis and calibration of data are given elsewhere⁷.

Eleven spots on seven individual apatite grains indicate that U concentrations vary significantly from 22.8 to 132 p.p.m. and do not show any correlation with $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios. A correlation diagram of $^{238}\text{U}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios of the Akilia apatites is shown in Fig. 1.

A least-squares fit using the York method gives the ^{238}U – $^{206}\text{Pb}^*$ isochron ages of $1,504 \pm 336$ (2σ ; mean square of weighted deviates (MSWD) = 6.4). A correlation diagram of $^{204}\text{Pb}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ ratios yields the $^{206}\text{Pb}^*$ – $^{207}\text{Pb}^*$ isochron ages of $1,459 \pm 160$ (2σ ; MSWD = 1.2). Both ages agree well with each other and are younger than the $3,860 \pm 10$ Myr of ref. 2 for BIFs. This suggests either that the apatites in the BIFs grew about 1,500 Myr ago, or that they grew earlier than that but were subsequently affected by recrystallization, and/or diffusive exchange with the environment, which reset the U–Pb system of the samples. If the apatites were formed during metamorphism, biogenic carbon could have been introduced during the event.

The Akilia association, including the BIFs and Isua supracrustals, are the oldest known components of the Archaean craton of Greenland and were affected by several metamorphic events after their formation 3,800 Myr ago (ref. 2). The latest event recorded in rock samples is the injection of basic dykes and crustally derived granitic sheets about 1,600 Myr ago, possibly coupled with the anatectic reheating⁸. Biotite from all types of gneisses in the area gives a Rb–Sr isochron age of 1,600–1,700 Myr ago (ref. 3), which is within the error of the isochron ages of the Akilia apatites.

If these apatites were formed 3,800 Myr ago, graphite inclusions within grains of apatites have also experienced a thermal event of around 600 °C about 1,500 Myr ago. The isotope composition of the carbonaceous inclusions might have been altered by the event. It has been suggested⁹ that the oxidation of carbonaceous matter during metamorphism could alter the initial ^{13}C value from –10‰ to –35‰ if temperatures are higher than 500 °C and if an oxidizing system is provided. In iron formation, an oxidizing system seems a reasonable suggestion.

The young U–Pb ages on the apatites, indicating closure for Pb in an event as late

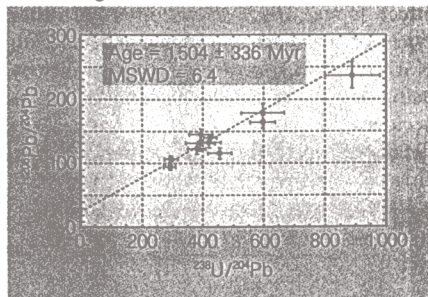


Figure 1 Correlation diagrams of $^{238}\text{U}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios of Akilia apatites. Errors are shown at the 1σ level. The dotted line shows the best fit by the York method. In the calculation of this $^{238}\text{U}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ diagram, an error correlation of $r = 0.8554$ was used, derived from the correlation coefficient between $\delta(^{238}\text{U}/^{204}\text{Pb})/(^{238}\text{U}/^{204}\text{Pb})$ and $\delta(^{206}\text{Pb}/^{204}\text{Pb})/(^{206}\text{Pb}/^{204}\text{Pb})$.

as 1,500 Myr, and the theoretical possibility that the light signature is a product of fractionation since 3,850 Myr, might in isolation throw doubt on the proposal¹ that the Akilia BIF hosted life when deposited at around 3,850 Myr. However, as stated by Mojzsis *et al.*¹, the petrographic association of the apatite and carbon-rich material is well known from unmetamorphosed BIF with microfossils, and also from modern biological observations. This was considered an equally important line of evidence for biological activity when the Akilia BIF was deposited.

In conclusion, when we seek evidence of the earliest life on Earth, we need to find an apatite with a U–Pb closure age apparently older than 3,500 Myr; with a pattern of rare-earth elements that indicates a biogenic signature; and with carbonaceous inclusions that contain isotopically light carbon.

Yuji Sano, Kentaro Terada,
Yoshio Takahashi, Allen P. Nutman

Department of Earth and Planetary Sciences,
Hiroshima University, Kagamiyama 1-3,
Higashi-Hiroshima 739, Japan
e-mail: ysano@ipc.hiroshima-u.ac.jp

- Mojzsis, S. J. *et al.* *Nature* **384**, 55–59 (1996).
- Nutman, A. P., McGregor, V. R., Friend, C. R. L., Bennett, V. C. & Kinny, P. D. *Precamb. Res.* **78**, 1–39 (1996).
- Baadsgaard, H., Lambert, R. St J. & Krupicka, J. *Geochim. Cosmochim. Acta* **40**, 513–527 (1976).
- Pankhurst, R. J., Moorbath, S., Rex, D. C. & Turner, G. *Earth Planet. Sci. Lett.* **20**, 157–170 (1973).
- Cherniak, D. J., Lanford, W. A. & Ryerson, F. J. *Geochim. Cosmochim. Acta* **55**, 1663–1673 (1991).
- Krogstad, E. J. & Walker, R. J. *Geochim. Cosmochim. Acta* **58**, 3845–3853 (1994).
- Sano, Y., Oyama, T., Terada, K. & Hidaka, H. *Chem. Geol.* **153**, 249–258 (1999).
- Kalsbeek, F., Bridgwater, D. & Boak, J. *Pap. Grønlands Geol. Unders.* **100**, 73–75 (1980).
- Eiler, J. M., Mojzsis, S. J. & Arrhenius, G. *Nature* **386**, 665 (1997).

