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# Preferential Dissolution of Radiogenic Pb from Alpha Damaged Sites in Annealed Minerals Provides a Mechanism for Fractionating Pb Isotopes in the Hydrosphere

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Simple laboratory (Hf) leaching experiments of zircon provide a clear link between enhanced solubility of $^{234}$U and radiogenic lead due to alpha-recoil damage (Davis and Krogh submitted; Mattinson 1994). Furthermore, because most upper crustal rocks cooled below annealing temperatures long after their formation, early formed lead rich in $^{207}$Pb is locked in annealed sites so that the leachable component is enriched in recently formed $^{206}$Pb. The isotopic composition of the leachable lead component then depends more on the cooling history and annealing temperatures of each host mineral than on their geological age; and the axiom that Pb isotopes cannot be fractionated in the natural environment, is invalid.

In our initial experiments designed to eliminate lead loss (discordance) from zircon populations, we found that selected high-quality zircons, after 15 minutes to 2 hours in cold Hf, exhibited increasing amounts of discordance but surprisingly had progressively higher $^{207}$Pb/$^{206}$Pb ratios (ages). On the hypothesis that the isotopic fractionation may reflect dissolution from alpha recoil damaged versus undamaged (annealed) sites, we carried out new tests monitoring $^{234}$U vs. $^{238}$U as well as radiogenic Pb.

Our first zircon sample was from an Archean trondhjemite for which 8 previously analysed fractions (4 abraded, 0.5 - 2% discordant, and 4 unabraded) defined an age of 2732±2 Ma with a lower intercept of 230 Ma. Analysis 1, carried out on abraded zircon without an Hf wash to test for secular equilibrium in the starting material, gave a near concordant result with a $^{234}$U anomaly of 0.47 ±0.19% only slightly below the accepted value for secular equilibrium. Analysis 2 of unabraded zircon washed in cold Hf for 1 hour and selected to be (white) alteration-free gave a result that was 8% discordant and 15 Ma older than the 2732 Ma age. Analysis 3 of abraded zircon that received a stronger leach (3.5 hours cold Hf, 1 hour warm Hf and held overnight in cold Hf) gave a residue that was 15% discordant with a $^{207}$Pb/$^{206}$Pb age more than 30 Ma older than the starting material, and a $^{234}$U anomaly of -11.0 ±0.4%. The wash had a positive $^{234}$U anomaly of +8.9 ±0.9%. The two data for the leached fractions project to 2728±5 Ma, within error of the accepted age but with a negative lower intercept of -749 +168/-139 Ma.

Our second test was carried out on zircons from a dacite for which 4 non-magnetic, abraded and unabraded zircon fractions (one concordant) gave an age of 2728±1 Ma with a lower intercept of 0 Ma. A single fraction given a strong leach, as per analysis 3 above, gave a residue that is 5% discordant, an age about 20 Ma older than the starting material and a $^{234}$U anomaly of -7.2 ±0.6%. The leachate gave a reversely discordant result with an age about 32 Ma too young and a positive $^{234}$U anomaly of +13.7 ±1.0%. The data are approximately collinear with that for the concordant abraded fraction through 2728 ±2 Ma but define a lower intercept of -1388 +147/-126 Ma. Clearly the lead isotopes are leachable along with $^{234}$U but the wash is enriched in recently formed $^{206}$Pb as if early formed 207-rich Pb (like $^{238}$U) is locked in annealed (undamaged) sites. A model relating the slope of the new discordia predicts a cooling time of 2500±20 and 2295±35 for the trondhjemite and dacite, respectively.

Although these experiments are based on a strong Hf attack on zircons, we believe, given the widespread $^{234}$U anomalies (of several hundred percent) observed in groundwater (Osmond and Cowart 1992), that they apply to the differential mobility of radiogenic Pb isotopes on a local and global scale. For example, Mississippi Valley type deposits believed to have been derived from leached upper crust by basinal brines may acquire their excess $^{206}$Pb by leaching of partially annealed uranium-bearing minerals. Given that the oceans have a positive $^{234}$U anomaly of 15% it is probable that they also receive a disproportionate amount of $^{206}$Pb over $^{207}$Pb compared to a continental source, and if amounts are sufficient, that this component could add excess $^{206}$Pb to the subduction-enriched upper mantle.

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Osmond JK & Cowart JB, *in Uranium Series Disequilibrium, Ivanovitch M & Harmon RS, eds. Clarendon Press, Oxford*, 290-333, (1992).