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Lead Retention in Zircons

The report by Gentry *et al.* (*1*) is incorrect on three important points. First, they are not the first workers to determine Pb isotope compositions of single zircon grains by mounting the grains directly on a mass spectrometer filament. This technique was successfully used more than 15 years ago (*2*), but has not been extensively pursued because of the limited usefulness of Pb isotope ratios alone for a mineral that almost always yields discordant U-Pb and Pb-Pb apparent ages (*3*). Furthermore, complete U-Pb isotope analyses are readily obtained for such samples by directly loading the residue from a high-pressure HF attack in the presence of a U-Pb spike (*4, 5*).

Second, as is well known by isotope geologists, the radiogenic $^{207}Pb/^{206}Pb$ of a mineral is very insensitive to Pb loss within the past 200 million years or so, due to the fact that ^{235}U, the parent of ^{207}Pb, has largely decayed by then. Thus, a 1500-million-year-old zircon could have lost 50 percent of its Pb 65 million years ago (a well-documented time of Pb loss in the southwestern United States) without lowering the $^{207}Pb/^{206}Pb$ by even 2 percent. In other words, a rough constancy of $^{207}Pb/^{206}Pb$ for zircons from the granite studied by Gentry *et al.* is not useful evidence that these zircons have lost little or no Pb. In fact, the data of Zartman (*6*) show that zircons from this rock at 2900 m depth have lost about 25 percent of their Pb. Moreover, the observation by Gentry *et al.* "that the total number of Pb counts per zircon . . . shows no systematic decrease with depth" in fact lends no additional support to their arguments. A simple linear regression of the data shows that they are much too scattered to provide any useful resolution of a trend.

Third, and perhaps most important, because natural zircons contain only trace amounts of radioactive elements, their Pb retentivity is not germane to the question of closed-system behavior of synthetic zircons that contain large amounts of highly radioactive nuclides. Such zircons would accumulate many orders of magnitude more radiation damage than natural zircons, with the potential for much greater elemental leachability. This point is of crucial importance to any element immobilization strategy (*7*).

Abundant data from geochronologic studies already exist to show that natural zircons can quantitatively retain Pb for up to billions of years (until a time of episodic Pb loss); otherwise zircon suites with concordant Pb/U ages or isotopic systematics indicating simple episodic Pb loss would not be observed.

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Ludwig *et al.* find no fault with our basic data (*1*) but claim to find other errors, to which I must take exception.

I acknowledge that Roubault *et al.* (*2*) previously analyzed single zircon grains, and we should have cited this in our report (*1*). However, in that instance the zircon was attacked by orthophosphoric and hydrofluoric acids after being placed on the filament. This procedure could have affected the lead isotopic measurements they reported, either directly from whatever lead that was in the acids or indirectly through molecular ion interferences generated by the acid treatment of the zircons.

The important point here is that their technique, which we learned of after our experiments were concluded, introduces the possibility of contaminant mass peaks in the lead region from two sources. In contrast, our report (*1*) describes an innovative mass spectrometric method which excludes any acid treatment of the zircon, and is thus a contamination-free technique.

As to the second point of Ludwig *et al.*, I note that their arguments about the lead ratios are in reality only problematical statements. Also, by agreeing that the total lead counts per zircon show no trend with depth, they fail to realize this is in fact strong evidence in favor of high lead retention. Significant lead loss would have been accompanied by a definite trend toward smaller total lead counts per zircon for the greater depths. But this was not observed. They also apparently fail to realize that their claim of a 25 percent lead loss from zircons taken from a depth of 9527 feet, which is based on a single zircon analysis cited by Zartman (*3*), is contradicted by their own admission that the total number of lead counts per zircon we obtained reveals no variation with depth. That is, a 25 percent lead loss at 9527 feet would imply an almost total lead loss (or equivalently few if any lead counts per zircon) at greater depths due to higher temperatures. As noted above, the data show no such effect.

In their third point, Ludwig *et al.* overlook the fallacy in their comparison of radionuclide retention in natural and synthetic zircons, namely, that the concentration of nuclear waste–type radionuclides in synthetic zircons can easily be adjusted so as not to exceed the naturally accumulated radiation dose during the required waste storage period. Thus, our results remain highly relevant to the solution of the long-term waste storage problem, a fact that continues to escape the Department of Energy but not the Congress (*4*).

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