Evidence of extinct ^{244}Pu in Hadean zircons

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The initial Pu/U ratio of the solar system is an important parameter in models of nucleosynthesis, mantle and atmosphere evolution and cosmochronology. Current estimates are based on analyses of the chondrite St Severin and the achondrite Angra dos Reis and are poorly constrained, ranging from 0.004 to 0.008. On account of its short, 82 Ma, half-life, ^{244}Pu was essentially extinct 3,900 Ma ago, and consequently there exists no reliable measurement of Pu/U for the Earth. Ages as old as 4.37 Ga have been documented for detrital zircons from the Jack Hills, Western Australia. The existence of zircons dating from the period when ^{244}Pu was 'live' offers an opportunity to measure the former terrestrial abundance of ^{244}Pu directly by way of fissiogenic xenon isotopes. The expected levels of fission xenon in individual zircons, (1 to 4 μg, 100 – 200 ppm U), are comparable to, the Xe blank levels (~10^{-15} ccSTP) typical of conventional noble gas mass spectrometers. To analyse these minute amounts of xenon we have made use of laser resonance ionisation using the RELAX mass spectrometer. We have carried out preliminary analyses of 3 individual 4,150 Ma zircons and one 3,600 Ma zircon from Western Australia, and obtained four clear fission spectra. All were free from significant atmospheric blank (^{130}Xe was less than 4x10^{-18} ccSTP, i.e. 100 atoms). The spectra of the older zircons clearly demonstrated the presence of varying proportions of ^{244}Pu fission xenon.

The highest ^{131}Xe/^{136}Xe, 0.134 ± 0.003, corresponds to an initial Pu/U ratio of 0.0035 ± 0.005. The variable ratios could result from loss of Xe after 4.0 Ga or represent U-Pu fractionation. We are currently analysing older zircons and intend to search for correlations with REE patterns, oxygen isotopes, and the degree of U-Pb concordance, and to investigate the thermal release characteristics of the xenon. In addition to constraining the terrestrial Pu/U ratio these investigations may allow us to characterise the geochemical behaviour of Pu in the earliest crust forming processes.