Pb isotopic analysis of individual fluid inclusions by LA-MC-ICP-MS: Progress over the past 60 months.

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Isotopic compositions of metals from fluid inclusions (FI) provide unique constraints on ancient sources and percolation paths of solute-rich aqueous solutions in the Earth's crust. Here we explore the analytical feasibility of Pb isotope ratio measurement in individual FI by LA-ICPMS. Synthetic FI in quartz of known Pb and TI isotopic compositions (SRM981 and SRM997 from NIST) were prepared at 700 °C / 1.8 kbar / 144 h in a cold-seal pressure vessel, with concentrations of 5000 $\mu g/g$ Pb, 1600 $\mu g/g$ TI, 10.5 wt-% NaCl and 5.9 wt-% KCl, respectively. Fluid inclusions in the run products were 10-30 μm on average, with a few reaching 80 μm diameter.

LA-ICPMS isotopic analysis was performed using a Nu Plasma or a Nu Plasma 1700 MC-ICPMS in transient data acquisition mode, and a Geolas 200Q laser system with computer-controlled stage. Raw data from SRM610 and FI shots were corrected for Hg interference (<0.5 % on mass 204), and exponentially for mass bias following the methods of Woodhead (2002) and Baxter et al. (2006).

Analyses on SRM610, used to bracket the FI analyses, yield within-run precisions as good as 100 ppm for isotope ratios normalized to ²⁰⁴Pb and 50 ppm for 207/206 and 208/206 ratios, respectively; 2 SD-uncertainties on shot-to-shot reproducibility per analytical session are as good as 400 and 200 ppm, respectively. Analyses of individual Pb-Tl Fl on both MC-ICPMS instruments vield accurate Pb isotopic ratios. albeit at poorer analytical precision (as good as 600 ppm 2 SE for 206/204, 207/204, and 208/204 and 400 ppm for 207/206 and 208/206). Inclusion-to-inclusion reproducibility is about a factor of 2-3 poorer. For analysis of Pb-bearing FI containing no TI, mass bias correction was achieved by admixing TI from a desolvating unit to the LA aerosol. Interestingly, Pb isotope ratios determined by this method are more precise and show a better inclusion-to-inclusion reproducibility than do the TI bearing FI, probably because of the presence of TI-rich "nuggets" in the Pb-TI FI, resulting in extremely fast, transient signals. Data on the Pb-only FI are accurate at the reported 2 SE level. Analysis of natural FI assemblages yield withinrun and inclusion-to-inclusion precisions even better than those obtained on the inclusion standards and demonstrate the usefulness of our analytical technique.

REFERENCES

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