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PhD Abstract

Pb-Pb step leaching (PbSL) is a digestion technique based on sequential acid treatment of a mineral resulting in the selective recovery of radiogenic and common Pb components from the crystal lattice, thereby making single-phase Pb-Pb dating possible. Developed originally by R. Frei and colleagues at Bern, the technique has had mixed success, but often the reasons for its failure have remained poorly understood.

One of the most difficult geological processes to constrain using existing geochronological methods is the timing of mineralisation in ore deposits, yet understanding when ore was formed is essential in the development of petrogenetic models. Some of the existing methods (e.g. Ar-Ar) are necessarily applied to alteration assemblages related to ore zones based on the assumption that both formed at the same time. In this study the PbSL technique has been applied to magnetite and sulfides (mainly chalcopyrite, but also pyrite, pentlandite/pyrrhotite) from a number of ore deposits (Mount Isa, Copper Blow, Ernest Henry, Osborne, Cadia-Ridgeway, Minnie Moxham and the Bushveld Complex) in order to assess the feasibility of the approach in constraining the ages of mineralisation from a phase that is unambiguously linked to the ore forming process. The choice of magnetite and chalcopyrite is based on the often surprisingly high U contents reported in several types of low-Pb sulfides and magnetite, yielding rather higher than expected initial U/Pb ratios (with ^{206}Pb/^{204}Pb up to 2000).

The majority of the Pb-Pb step-leaching arrays produced in this study are highly scattered and generate younger than expected age estimates with large uncertainties. The lack of agreement between the data obtained from this study and the generally ‘accepted’ ages for mineral deposition at these deposits is best explained by open-system U-Pb isotope behaviour. Additional potential sources of scatter in the ‘isochrons’ include: insufficient U contents in the samples to allow adequate ingrowth of radiogenic Pb; excessive concentrations of Pb that shield the radiogenic signature; and a lack of spread in ^{207}Pb/^{204}Pb arrays for young samples (as the majority of ^{235}U decayed early in the Earth’s history).

In this study, the combination of PbSL and U/Pb geochronology with solution-based trace element ICPMS, SEM element mapping and *in situ* LA-ICPMS Pb isotope analysis has been employed. Together, these provide a powerful approach for revealing and understanding the details of open-system U/Pb behaviour. Age estimates obtained from U/Pb geochronology are shown to be inconsistent between samples and within aliquots of the same sample indicating small-scale U/Pb heterogeneity. ICPMS trace element analysis, LA-ICPMS and SEM imaging reveal U-rich impurities on a range of scales. Investigations with SEM back-scattered imagery reveal a significant population of U-rich impurities (~3% of the grain) in only one of the magnetites from this study despite trace element ICPMS results that infer the presence of U-, Ca-, Ba- or Zr-rich impurities in the majority of the samples. The release patterns of Ca and Ba often correlate with U indicating co-siting of these elements. This infers that the bulk of radiogenic Pb is likely to have been sourced from these impurities rather than lattice sites within the host phase. LA-ICPMS analyses provide further evidence for the presence of sub-microscopic U-rich impurities, with linear traverses of magnetite producing well-pronounced ^{206}Pb-rich data 'spikes' against a low ^{206}Pb/^{204}Pb background.

Despite the complexities in U-Pb behaviour unravelled in this investigation, it would appear that many of the 'isochrons' do provide some evidence for the timing of isotopic disturbance. In the case of the Curnamona and Isa provinces, the PbSL results are broadly consistent with a ~1.1 Ga 'age' and therefore would appear to reflect the major tectonic reorganisations and mafic magmatism believed to have occurred at around this time.

This study highlights the need for caution when applying PbSL (or U-Pb) geochronological techniques to ore minerals. Even in cases where an 'isochron' appears to be well constrained, it is possible that the age is spurious owing to open system behaviour. It has been possible to detect such complexities here by the choice of examples (i.e., from relatively well-constrained deposits were independent age information is available) and the comprehensive analytical approach employed. In cases where independent constraints are unavailable, and detailed geochemical analysis is not performed, there is some danger that the 'ages' produced and reported do not reflect those of the events they are believed to represent. A new approach is proposed in order to help assess the veracity of PbSL age information in such cases.