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Review

The oxygen isotopic composition of uranium minerals: A review

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ABSTRACT

Uranium ore is an essential material in the preparation of nuclear fuel for civilian as well as military uses. Uranium is first extracted from uranium-bearing minerals using a variety of reagents, and is precipitated from solutions as yellow cake prior to isotope enrichment processes. The disintegration of the former Eastern Bloc in the 1990s and frequent unrest in the Middle East have underscored the need for better characterizing source uranium ores for forensic and attribution purposes.

The world's major deposits of U occur in several distinctly different geological environments. Fourteen principal types of U deposits and rocks with elevated uranium contents are recognized with more than 40 subtypes. Combining our own analysis and literature data, we have amassed over 250 oxygen isotope data from 13 major U-producing countries, which vary widely from −32 to +11‰. However, interpreting the oxygen isotopic composition of uraninite in terms of the composition of the fluid from which it precipitated, or interacted with, requires knowledge of the fractionation factor and temperature of interactions, which are not always available. Since each deposit type occurs within a unique geologic setting and is generally formed from chemically distinct fluids, the chemical compositions of the uranium ores are also distinct: uranium deposits that form in igneous rocks have higher trace element compositions relative to sandstone-hosted deposits. Our data shows that one of the most useful techniques for distinguishing between uranium ore is to use a combination of $\delta^{18}O$ values and rare-earth elements (e.g., La/Yb ratios). These methods may allow investigators to trace uranium ore back to the source.

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1. Introduction

Uranium is a trace element in the crust (2.7 ppm in the upper crust and 1 to 1.7 ppm is the global crustal average) and mantle (~0.015 ppm) and is often found in accessory minerals such as zircon. However, high concentrations of U do occur on many continents throughout the world (Fig. 1) where U deposits can contain millions of kilograms of U (e.g., unconformity-type U deposits, Canada) and, under the appropriate conditions, the U concentration and appropriate $^{235}U/^{238}U$ ratio was sufficient to sustain fission chain reactions (e.g., Oklo, Gabon). Uranium deposits are also important sources of energy for countries that rely heavily on nuclear power (e.g. France), and they have been used to model Earth's ancient climates (e.g., Holland, 1984) and as natural analogs for the geologic disposal of radioactive waste (e.g., Gauthier-Lafaye et al., 1996).

The mining of uranium ore is an essential step in the preparation of nuclear fuel for civilian as well as military uses. In general, uranium is extracted from rocks containing uranium-bearing minerals (ore) using a variety of reagents. Once the uranium is in solution, it is precipitated as yellow cake using a number of precipitants including ammonia, magnesium oxide, sodium hydroxide, and hydrogen peroxide (e.g., Dunworth, 1963; Gupta et al., 2004; Jackson, 1981; Merritt, 1971).

The oxygen isotopic composition of UO_2 can potentially be used to fingerprint uranium ore (Linares, 2006; Pajo, 2001). Interpreting the oxygen isotopic composition of uraninite in terms of the composition of the fluid from which it precipitated, or interacted with, requires knowledge of the fractionation factor and temperature of interaction. However, since each deposit type occurs within a unique geologic setting and is generally formed from chemically distinct fluids, the chemical compositions of the uranium ores are also distinct. For example, intrusive or igneous rocks generally have elevated trace element compositions relative to sedimentary rocks. Consequently, uranium deposits that form in igneous rocks have higher trace element compositions relative to sandstone-hosted deposits. Therefore, the trace element composition of the uranium ore is potentially a powerful tool to distinguish between ore mined from different deposits. Here we present a global compilation of the $\delta^{18}O$ values of uranium minerals from numerous deposit types and show that the $\delta^{18}O$ values and rare earth element (REE) chemistry of uranium ore can be used to fingerprint uranium ore.

2. Uranium deposits

2.1. Uranium geochemistry and mineralogy

The geology, mineralogy and geochemistry of uranium deposits have been described in numerous papers and books (e.g., Burns and Finch, 1999; Cuney and Kyser, 2009a; Cuney, 2009, 2010; Dahlkamp,

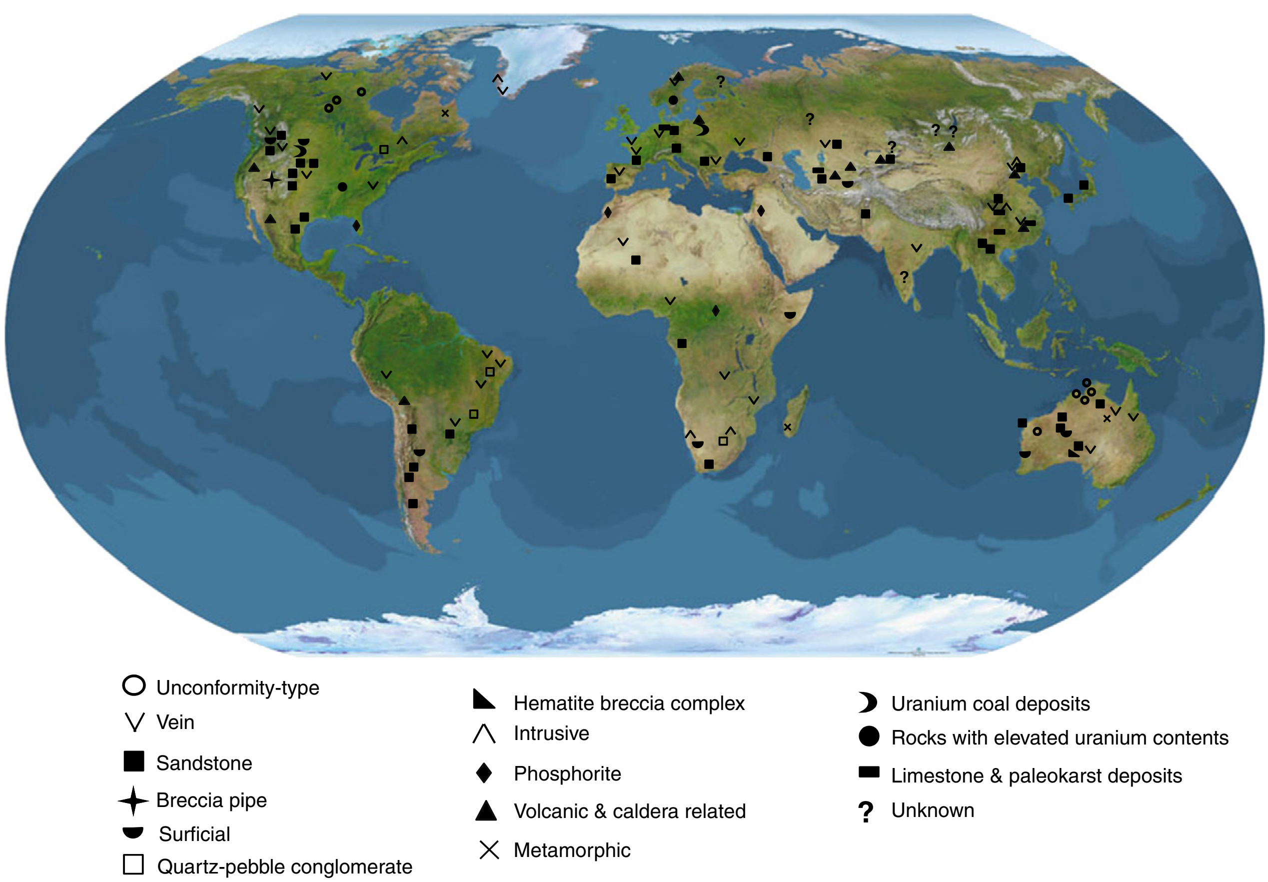

Fig. 1. Global distribution of selected uranium deposits.
Data are from Dahlkamp (1993) and Finch et al. (2005).