

The degradation of monazite: Implications for the mobility of rare-earth and actinide elements during low-temperature alteration

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Abstract: Monazite is an important economic source of thorium, the rare-earth elements and uranium. This and its chemically inert nature has led to speculation that artificial phosphate-based matrices similar in composition to monazite may prove useful as wasteforms for high-level radioactive waste. In order to assess the long-term degradation behaviour of monazite, an integrated geological, hydrogeological and mineralogical study was undertaken at the Steenkampskraal monazite mine, South Africa. Steenkampskraal is among the richest monazite ore bodies in the world comprising up to 45 weight % rare-earth oxides, 8.8% thorium oxides and 600 ppm uranium. Optical and electron-microprobe analyses of the ore reveal distinctive alteration patterns with uranium appearing to be lost preferentially. The heavy rare earths are also preferentially removed from the ore, giving rise to a marked fractionation in altered grains. Thorium, although leached, is re-concentrated together with the heavier rare-earth elements in microcrystalline silicate and oxide alteration products within the host rock. The implications for waste encapsulation are discussed from the perspective of potential groundwater transport away from the source.

Key-words: rare-earth elements, thorium, uranium, actinides, monazite alteration, Steenkampskraal, South Africa.

Introduction

The geochemical stability of natural lanthanide minerals, such as monazite, xenotime and allanite, all of which contain some thorium and uranium, is of considerable relevance to radioactive waste disposal for two main reasons. First, it has encouraged research into analogous synthetic phases incorporating artificial isotopes (*e.g.*, Bénard *et al.*, 1996; Dacheux *et al.*, 1998). Secondly, establishing the fate of U and Th released during mineral alteration processes provides the only means of verifying long-term predictions of actinide migration away from an engineered repository (Read & Hooker, 1992).

Monazite [(La,Ce,Nd,Th)PO₄] is one of the principal sources of rare-earth elements (REE) and thorium in the continental crust. Economically viable concentrations occur in placer deposits and as massive ore bodies in igneous and metamorphic provinces, as is the case at Steenkampskraal, South Africa (Fig. 1). Steenkampskraal is thought to be the richest monazite ore body in the world comprising up to 45 weight % REE oxides, 8.8% thorium oxide, 18% P₂O₅ and 600 ppm uranium. In addition to these elements, Steenkampskraal contains more than 1 weight % copper, up to

1.5% zirconium and traces of gold (Pike, 1959). Locally, the ore shows enrichment in iron and titanium oxides in common with other mineral deposits in the region. The mine was first in operation from 1952 to 1963 when it produced an estimated 50,000 tonne of ore, and ranked as the world's leading source of thorium and rare-earth elements. Surface spoil heaps, presently a source of environmental contamination, are likely to be processed should the mine be re-opened at some time in the future

This paper describes the petrography and mineral chemistry of monazite from Steenkampskraal and examines new evidence concerning the alteration that it has undergone. The latter aspect is of particular interest because, owing to its well-known persistence in nature, published work on the degradation of monazite is scarce. Poitrasson *et al.* (1996) give an account of two potential mechanisms for the hydrothermal alteration of monazite in granites from the United Kingdom, where the processes of chloritisation and greisenisation are believed to result in distinct REE patterns. More recently, Hecht & Cuney (2000) have reported evidence of monazite alteration in the Precambrian basement of the Athabasca Basin, again under hydrothermal conditions. Other studies carried out in this area (*e.g.*, Alderton *et al.*,