The significance of Uranium and Thorium concentrations in pegmatitic leucogranites (alaskites), Rössing Mine, Swakopmund, Namibia

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Studies completed on the Damaran pegmatitic leucogranites, intruded as dykes and sheets in the vicinity of the Rössing Uranium Mine, indicate that they are derived from the residual crystallization products of a late Damaran granitic pluton intruded into the lower formations of the Damaran cover rocks transecting an earlier ductile shear zone generated during orogenic transpression. These structural pathways have acted as controlling conduits for later transtensional granite emplacement and uranium mineralization. The fluid-rich pegmatitic leucogranites in the cover rocks are therefore considered to be fractionated products of granitic magma, generated in a transtensional environment as a consequence of orogen extensional collapse following oblique collision between the Congo and Kalahari cratons. This transtensional tectonic setting generated a wide range of leucogranite normative compositions, but the majority of the leucogranites at the Rössing Mine are pegmatitic alkali-leucogranites (alaskites) exploited economically for their uraninite and beta-uranophane. All the Rössing leucogranites have anomalously low Th/U ratios. The mineralization at the Rössing Mine can be viewed as successive concentrations of U relative to Th related firstly to a primary magmatic process of pegmatite formation; secondly to hydrothermal redistribution, and finally to supergene enrichment. The tripartite series of geochemical processes has caused the decoupling of uranium and thorium concentrations so that U is considerably enriched relative to Th in the ore reserves used for mining. Such natural geochemical changes have considerable benefits to the workforce at Rössing. Health hazards from uranium and thorium are not as serious as those found in other uranium deposits where Th/U ratios are closer to the natural crustal abundance of 4 to I.

Introduction

In 1992, Dropkin and Clark published a document questioning the health and environmental risks at Rössing Uranium Mine. Because of the seriousness of the allegations the Government of Namibia requested that the International Atomic Energy Agency should send radiation experts to Namibia to assess the safety, health and environmental risks at Rössing Uranium Mine. The IAEA invited the International Labour Organisation and the World Health Organisation (represented through the Ministry of Health and Social Services of Namibia) to take part in this Technical Mission. The Terms of Reference for the IAEA Technical Mission, which took place from 31 August to 11 September 1992 were :-

a) To corroborate the results of radiation monitoring so far carried out by Rössing by making independent measurements.

b) To carry out an assessment of the planned programme for the management of uranium tailings, including decommissioning and rehabilitation of the tailings pile.

c) To assess the general occupational safety, including engineering and operational safety and working conditions at various facilities of the Rössing Mine and mill-complex.

d) To make an assessment of the long-term health effects from exposure to radiation received.

The IAEA Report (Ahmed *et al.*, 1993) on the radiological health and safety situation at Rössing indicated that uranium mining in Namibia is a low risk operation. The Technical Mission also commented that the radiation exposure levels at various sites in the Rössing Uranium Mine and throughout the processing plant were very low, much lower than current international limits. Despite these comments Dropkin (1993) continued to question the health hazards of uranium mining. However, the environmental question that should be asked is:-

"Why are the radiation exposure levels within the Rössing Mine relatively low, and not a serious health hazard?".

This paper attempts to provide a geochemical explanation based on recent field work and geological studies in the Central Zone of the Damara Orogen, western Namibia in the Khan River Gorge, the Rössing Dome and at the Uranium Mine.

Tectonic setting

The commonly held view of the relationship in the Central Zone of the Damara Orogen between the Neoproterozoic metasedimentary cover rocks and the underlying red granite gneiss in western Namibia, was that the red granite gneiss represented syn-tectonic granite intruded and deformed during the Damara Orogen (Smith, 1965; Miller, 1983). This hypothesis was challenged by Kröner *et al.* (1991) who suggested that the red granite gneiss could be Grenville in age (1040 Ma) and at least 300 My older than the overlying Damaran metasedimentary cover. Furthermore, the syn-tectonic red granite (gneiss) was interpreted by Smith (1965) as intruding the Abbabis 'basement'. Recent structural mapping by Oliver (1993) has shown that a significant high strain zone of *L-* and *L/S* tectonites can be traced within the overlying Etusis Formation of the Damaran metasedimentary sequence as well as in the red granite gneiss below. This means that the whole zone consisting of the basal unit of the Etusis Formation and the red granite gneiss can be interpreted as a ductile shear zone. Structural criteria (Oliver, 1993) indicate that the Damaran cover has been detached and translated towards the southwest.

Structure and composition of the Damaran cover

Regional mapping by Smith (1965) suggested that the Damaran cover is domed by granite and infolded into tight synclinal keels between the domes. According to Oliver (1995) the folding is the consequence of initial compression and crustal thickening of Damaran metasedimentary cover. The domes are thought by Oliver (1995) to be the result of combined crustal N-S transpression and SW-NE extension.

A recent appraisal of fabrics in the cover rocks of orogens by Dewey (1995) suggests that kinematic field measurements indicating constriction should be regarded as products of transtension in the central part of the orogen. This new concept coupled with the known clockwise rotation of the Kalahari craton and the subsequent movement of the Congo craton to the northeast, during collision of West and East Gondwana, invokes transtensional extension in the central part of the Damara Orogen coincident with thrusting onto the craton margins (Dewey, 1995). This latter period of transtension is coeval with late Damaran granitic plutonism witnessed by the emplacement of granitoids ranging in composition from granodiorite to alkali granite (Bowden and Tack, 1995). The pegmatitic leucogranites at Rössing constitute part of the late Damaran transtensional plutonism.

The cover rocks are dominantly unmelted and metasedimentary with minor basic meta-igneous suites (at cordierite-garnet-K-feldspar grade, characteristic of the ductile middle crust). They are currently described in terms of a series of discrete stratigraphic formations (Miller, 1983). Observations show that the lower sequences of the Etusis Formation are mylonitic, while high strain zones within the Khan and the Rössing Formations possibly take the form of linked shears that cut down to the base of the cover. Stretching directions in mixtite pebbles of the Chuos Formation also support

this view of high strain. The balance of evidence suggests that not only is the cover sequence allochthonous but also that regionally, significant strain zones exist within the Damaran cover which were subsequently exploited as pathways by the late Damaran, transtensional, fluid-rich granitic magmas.

Damaran granitic rocks

Some of the granitic rocks found throughout the Damara Orogen (Miller, 1983) display a distinctive mineral fabric most probably related to a tectonic overprint generated during transpression. The mineralogical and chemical compositions of these transpressional granitoids are typical calc-alkaline. In contrast the later transtensional granitoids have a more distinctive alkaline signature (Mestres-Ridge, 1992) characteristic of granitic sheets in the Damaran cover on the southern side of the Rössing Dome at the Rössing Uranium Mine. The range of normative compositions are presented in Fig. 1. The broad range is typical for leucogranite compositions in orogens (Lameyre and Bowden, 1982) varying from granodiorite, through monzogranite, and dominated by syenogranite to alkali granite. At Rössing, only the coarser grained syenogranite and in particular the pegmatitic alkali-leucogranite contain primary uraninite as an economic accessory mineral.

The pegmatitic leucogranites

The leucogranites have been studied in the cover rocks particularly in the vicinity of the Uranium Mine on the southwestern flank of the Rössing Dome (Mestres-Ridge, 1992). Here the leucogranites are characteristically pegmatitic and outcrop as a series of sheets particularly concentrated as curving swarms following the folds in the Khan and Rössing Formations along the flanks of the Rössing Dome. The pegmatitic leucogranite sheets in the cover rocks are oriented NE-SW along the length of the orogen. Whole rock Rb-Sr dating for samples from the Rössing Uranium Mine give two groups of ages 510 Ma (Kröner, 1982), and 458 Ma (Hawks worth *et al.*, 1983) with contrasting initial strontium ratios. This suggests that the granite plutonism peaked around 510 Ma although Rb-Sr mineral blocking temperatures did not close until ~450 Ma suggesting either repeated pulses of granite intrusion and/or prolonged hydrothermal activity.

In the Khan River Gorge the leucogranites are seen to passively invade, without any significant deformation, the metasedimentary cover rocks as structurally controlled sheets, feeding plugs, stocks and bosses trapped beneath marble horizons. New radiometric U-Pb, Rb-Sr and Sm-Nd methods are currently in progress to define precisely the time of emplacement, the hydrothermal history for the leucogranites, and the source region for magma generation.
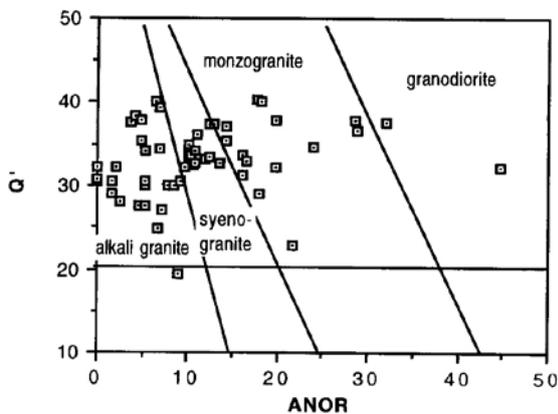
Figure 1: Normative compositions of Damaran granitic sheets within the cover formations at the Rössing Uranium Mine.
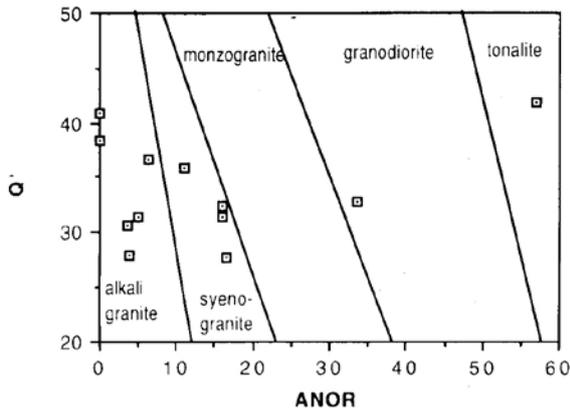
Figure 2: Normative compositions for Damaran granitic sheets intruding the base of the cover in the Khan Gorge and Ida Dome.
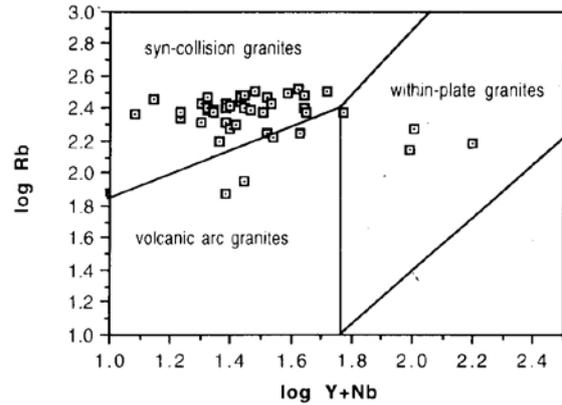
Figure 3: Tectonic settings (Pearce *et al.*, 1984) for Damaran granitic sheets intruding the cover Formations.

Geochemical and petrogenetic constraints on the origin of leucogranites

Recent geochemical studies by Mestres-Ridge (1992) indicate that the pegmatitic alkali-leucogranites at Rössing have a distinctive A-type granite signature (Sylvester, 1989). However, the alkali-leucogranites (enriched in uraninite) should be regarded as fractionated products from a parental magma rich in K evolving from diorite/quartz monzodiorite - quartz monzonite compositions (Mamood and Cornejo, 1992) typical of Pan-African plutons elsewhere in Africa (Liegeois *et al.*, 1994). Such a wide range in rock types is supported by the normative compositions for Damaran granitic sheets intruding the base of the folded cover formations in the Khan Gorge and Ida Dome (Fig. 2) and by the incomplete tectonic discrimination shown by trace elements (Fig. 3).

Significantly, the majority of the granitic sheets in the cover rocks at the Rössing Uranium Mine can be regarded as fractionated I/S types (Fig. 4). However, some caution should be applied to the use of Figs 3 & 4 particularly if the HFS elements are mobile under hydrothermal conditions.

Field work suggests that leucogranite magma(s) invading the Damaran cover rocks used existing structural features as conduits, mimicking these structures and impounding against rheological barriers. The pegmatitic nature of the leucogranites indicates that they were fluid rich products of substantial volumes of fractionating granitic magmas. Hence a large granite pluton, or a series of linked plutons as a major batholith of Pan-African age must have fractionated, slowly cooled, and crystallized below the mineralized Damaran cover rocks (Bowden and Tack, 1995).

The favoured hypothesis, based on field work in Namibia (Oliver, 1993, 1995) and West Africa for the Pan-African orogeny (Black and Liegeois, 1993), is that the granitic magmas were generated as a consequence of orogenic extension following collision between the

Congo and Kalahari cratons. Transpressional collision first induced thickening of both crust and the mechanical boundary layer (MBL) of the continental lithospheric mantle. Thermal instability (Bird, 1979) of the MBL plunged into hot as the nosphere induced delamination of the lithospheric mantle. This was accompanied by hot as the nosphere upwelling (Molnar, 1988), and fusion of the lower crust (Black and Liegeois, 1993) causing transtensional extension. This process was responsible for the generation of large volumes of granitic magma(s) with initial crustal signatures. With continued uprise of the as the nosphere the crustal melts interacted with mantle components producing transtensional granitoids. The transtensional granitic magmas with their volatile fluxes were emplaced as plutons crosscutting the high strain zones at the base of the Damaran cover and ponded at rheological interfaces beneath and within the Damaran cover producing a widespread thermal metamorphic overprint. A coeval hydrothermal overprint generated from the cooling granite pluton(s) affected the cover rocks and the exogenic granite sheets, modifying their original geochemical signatures, and generating significant uranium mineralization in the
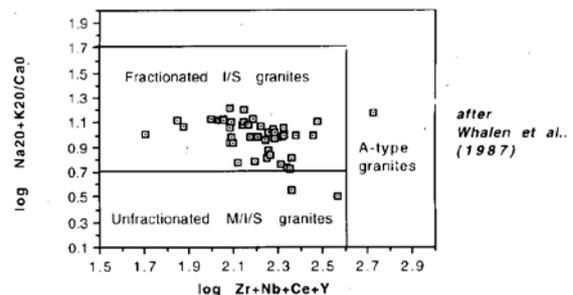
Figure 4: Granite classification for Damaran granitic sheets in the cover formations at the Rössing Uranium Mine (after Whalen *et al.*, 1987)

marginal zones between the pegmatitic leucogranite sheets and the Damaran cover.

Uranium geochemistry

Uranium (atomic number 92) is found naturally as U^{238} (99.3%) and U^{235} (0.7%). It can be considered as the fourth member of a series of elements (the actinides) which resembles the rare earths (the lanthanides). Uranium is a lithophile element which is enriched in the crust (2 ppm). In nature uranium normally occurs as tetravalentions (U^{+4}) with hexavalent ions (U^{+6}) Stable only under oxidising conditions. Its ionic potential indicates that it does not readily substitute in crystal lattices of normal rock-forming minerals except in trace amounts. However, significant quantities of uranium are frequently found in accessory minerals such as thorianite, thorite, thorogummite, allanite, xenotime, zircon, fluorite, apatite and barite. Much of the uranium in rocks is loosely held mainly as film coatings on rock-forming minerals which can be easily leached with dilute acids.

Under oxidising conditions, leaching of uranium from weathered outcrops is rapid and is enhanced by the formation of complex uranyl ions with carbonate and sulphate anions. Uranium can also be removed from oxidising solutions by reduction in the presence of sulphides. Furthermore uranium can be absorbed on clay minerals, and iron hydroxides such as goethite.

Uraninite is the most abundant uranium mineral as U^{+4}. Magmatic and pegmatitic uraninite contain substantial amounts of thorium and the rare earths. In contrast, hydrothermal uranium deposits only have low concentrations of Th and REE.

Uranium mineralization in western Namibia

With the discovery of high strain zones in the Damaran cover in an area of significant uranium mineralization it is possible to conclude that leucogranite magma and hydrothermal fluid utilised these structural zones as important channelways. Previously reported U-Pb dating on uraninite from pegmatitic leucogranites at Goanikontes (509 ± 1 Ma) suggest that leucogranite magmatism and primary uranium mineralization were contemporary (Briqueu *et al.*, 1980).

Around the Rössing Dome, the Damaran cover sequence, composed of mylonitised Etusis Formation and overlying subsidiary shear horizons in the Khan and Rössing Formations, were particularly favourable for the intrusion of pegmatitic U-rich leucogranites (alaskites). The swarms of pegmatitic leucogranite are locally so intense around the margins of the dome, that the Damaran host rocks only outcrop as trapped enclaves between successive multiple granitic sheets.

At the Rössing Mine the pegmatites consist largely of anhedral smoky quartz, subhedral salmon-pink microcline, white albite, and accessory black biotite. They are intruded as multiple sheets and dykes associated with aplite utilising the high strain zones as conduits and following the fold structures in the Damaran cover rocks.

Primary uranium mineralization at the Rössing Mine

The major 'uranium mineral at Rössing (55%) is uraninite, closely associated with zircon, apatite, titanite and monazite. The uraninite occurs as minute grains (0.3 mm in diameter) either as inclusions or along cracks in quartz, feldspar, and biotite or as free interstitial grains in pegmatite.

Hydrothermal uranium mineralization at the Rössing Mine

Evidence for hydrothermal processes can be demonstrated by disequilibrium mineral assemblages in the granitic sheets as well as the formation of Cu-Fe-Mo-As-S ore minerals. For example, the mineral betafite separated from the granitic sheets at the Rössing Uranium Mine shows a wide range in composition as well as containing partial breakdown products as a result of changing fluid PT conditions. The average concentration of betafite does not exceed 5%. Minor pyrite, chalcopyrite, bornite, molybdenite, arsenopyrite, ilmenomagnetite, fluorite, and hematite are associated with the formation of hydrothermal betafite.

Hydrothermal uraninite can be recognised by the unusually low concentrations of the rare earths (REE) and thorium (Rich *et al.*, 1977). Mestres-Ridge (1992) has indicated that the whole-rock rare earth concentrations in leucogranite samples from an area adjacent to the mine are substantially depleted in total REE. Furthermore, the Th/U ratios in the Mestres-Ridge samples are anomalously low.

As well as the crystallization of hydrothermal betafite and uraninite, alkali feldspars in the granitic sheets are significantly ordered with triclinicities close to 1. The ordering of alkali feldspars in response to pervasive hydrothermal fluids has been well established (Bowden and Kinnaird, 1984; Bowden *et al.*, 1987). Likewise beta-quartz readily re-equilibrates and recrystallizes to alpha-quartz trapping hydrothermal fluids as fluid inclusions. Recent studies suggest that the dark smoky anhedral quartz crystals, formed at the contact margins of the granitic sheets, contain trapped fluid inclusions relatively rich in non-condensable gases such as nitrogen and methane, as well as varying proportions of carbon dioxide and low salinity aqueous solutions. NMR studies on the same quartz crystals indicated that hydroxyl molecules are present in the trigonal mineral structure (Ratcliffe, 1991)

These observations suggest that the granitic sheets in the Rössing uranium deposit should be recognised as exhibiting significant hydrothermal overprinting of the original magmatic mineralogical and geochemical sig-

natures (Kerr, 1990).

Supergene uranium mineralization at the Rössing Mine

In modem times the sheared horizons act as water aquifers redistributing secondary uranium minerals along minor faults, joints and sheared contacts within the cover rocks. At Rössing approximately 40% of the uranium deposit consists of secondary supergene minerals. These include beta-uranophane, metatorbernite, metahaiweeite, uranophane, carnotite, thorogummite, and gummite. A major part of the supergene mineralization forms mineral coatings along joints, cracks, contact zones, fault planes, subsidiary shears, as well as penetrating mineral cleavages, and grain boundaries in quartz and feldspar, and between flakes of biotite. Particularly significant are the enriched uranium concentrations along faulted contact zones between leucogranite sheets and Damaran country-rocks generated as a consequence of supergene mineralization.

Thorium geochemistry

Thorium (atomic number 90) is the second member of the actinide series of elements. It commonly occurs as Th^{232}. The most common oxidation state is Th^{+4}, and its compounds resemble those of tetravalent cerium (lanthanides), and to some extent, Ti, Zr and Hf.

Geochemically Th behaves differently than U particularly in hydrotheffilal and supergene mineralization processes. Whilst U is far more mobile because of its increased solubility at higher oxidation states, Th remains locked into the lattice sites of pegmatitic minerals crystallised from leucogranite magmas.

Uranium and thorium at the Rössing Uranium Mine

Geochemical investigations of uranium and thorium concentrations in pegmatitic leucogranites support the observations of Ruzicka (1975) that supergene uranium represent an important component in the enriched uranium deposit at Rössing. This is reflected in the unusual Th/U ratios presented in Fig. 5. It is suggested that the Th/U ratio of 0.39 represents hydrotheffilal mineralization, and supergene uranium enrichment marked by a Th/U ratio of 0.08. The original Th/U magmatic signature is virtually eliminated by hydrotheffilal and supergene processes and cannot be easily displayed on Fig. 5. Such natural geochemical changes have considerable benefits to the workforce at Rössing. Health hazards from uranium and thorium are not as serious as those found in other uranium deposits where Th/U ratios are closer to the natural crustal abundance of 4 to 1.

Health hazards

Thorium and particularly uranium are toxic in the same manner as other heavy metals such as lead, copper, arsenic or antimony. Superimposed on this toxicity, however, are other hazards associated with the radioactive decay of uranium and thorium. Both elements are members of decay series in which alpha, beta and gamma radiation are emitted, and boili are responsible for the evolution of radioactive gases, radon and thoron, respectively. These gases deposit solid radioactive substances when they decay, and it is clearly necessary to ensure that this deposition does not take place inside the lungs or other parts of the body.

According to the UK Ionising Radiations Regulations (1985), thorium is more dangerous to the human body than uranium. Airborne contamination for uranium should not exceed 0.2 Bq per cubic metre of air, but thorium should not exceed 0.01 Bq per cubic metre of air. Contamination from dust particles is such that natural thorium is ten times more dangerous than natural uranium.

However with such low Th/U ratios in the Rössing Uranium Mine, and low level of exposures, the probabilities of radiation-induced occupational illnesses are extremely small, and well within acceptable levels of risk in safe industries. Furthermore, the IAEA Technical Mission (Ahmed *et al.*, 1993) found that the management of the mill tailings at Rössing, and their associated tailings surveillance programme were of a good standard and conformed to current international standards.

Conclusions

Studies completed on the Damaran pegmatitic leucogranites, intruded as dykes and sheets in the vicinity of the Rössing Uranium Mine, indicate that they are sourced from a late Damaran granitic pluton along structural pathways in the lower formations of the Damaran cover rocks. These pathways have acted as controlling conduits for granite emplacement and uranium mineralization. The fluid-rich leucogranites in the cover rocks are considered to be fractionated products of granitic magmas, generated in a transtensional environment as
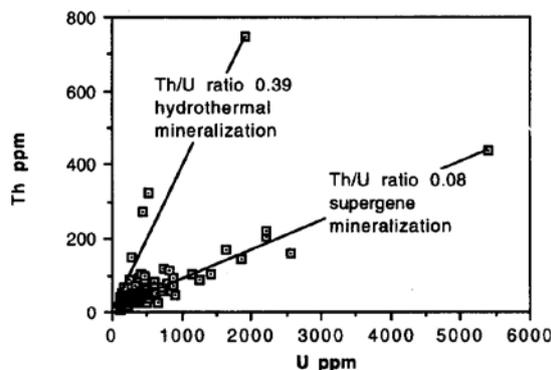
Figure 5: Variations in the Thorium-Uranium ratio for Damaran granitic sheets at the Rössing Uranium Mine.

a consequence of orogen extension following oblique collision of the Congo and Kalahari cratons.

There is a wide range of leucogranite normative compositions, but the majority of the leucogranites at the Rössing Mine are pegmatitic alkali-leucogranites (alaskites) exploited economically for their uraninite and beta-uranophane. All the Rössing leucogranites have anomalously low Th/U ratios.

The uranium mineralization at the Rössing Uranium Mine can be viewed as successive concentrations of uranium relative to thorium related, firstly, to a primary magmatic process of pegmatite formation; secondly, to hydrotheffilal redistribution, and finally, to supergene enrichment. The tripartite series of geochemical processes has caused the decoupling of U and Th concentrations such that

U is considerably enriched relative to Th in the ore reserves used for mining. Such natural geochemical changes have considerable benefits to the workforce at Rössing. Health hazards from uranium and thorium are not as serious as those found in other uranium deposits where Th/U ratios are closer to the natural crustal abundance of 4 to 1.

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