The Source of Witwatersrand Gold: Evidence from Uraninite Chemistry

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Abstract. An in-situ LA-ICP-MS study of different generations of uraninite from the Mesoarchaean Witwatersrand gold palaeoplacer deposits revealed unusually high Au concentrations in rounded, detrital uraninite grains but no detectable Au in secondary, hydrothermally mobilised uraninite. A Au-enriched uraninite-bearing magmatic host is suggested as a significant source for detrital gold in the Witwatersrand sediments.

Keywords. Uraninite, trace elements, gold, Witwatersrand

1 Introduction

Hardly any other gold province has been studied and debated so thoroughly as the Witwatersrand Basin in South Africa. A vast range of genetic models have been suggested to explain the unique accumulation of gold in the Meso- to Neoarchaean sediments there, ranging from placer, magmatic, metamorphic, various syn- or post-depositional hydrothermal models to impact-induced, syn-sedimentary microbially mediated formation to precipitation from seawater and hydrogen degassing of the Earth's core. Over the past decade consensus has emerged amongst most workers that the auriferous conglomerates of the Witwatersrand represent palaeoplacer deposits with most, if not all, of the gold having entered the host conglomerates as detrital particles that subsequently experienced some short-range mobilisation by post-depositional fluids (for a review and further references see Frimmel et al. 2005). In spite of significant progress in our understanding of the genesis of the Witwatersrand deposits over the past years, the ultimate question of the source of all this gold remains unanswered.

The clast lithology and the detrital mineralogy of the gold-bearing metasedimentary rocks indicate both a felsic and mafic to ultramafic provenance comparable with the kind of granitoid-greenstone terrane that makes up most of the pre-Witwatersrand rocks in the Kaapvaal Craton. Many of those who advocated a palaeoplacer model in the past sought the source of the gold in greenstone-hosted, orogenic-type gold-quartz veins as currently mined, for example, in the Barberton greenstone belt (e.g. Robb & Meyer. However, more than two decades ago Hallbauer & Barton (1987) pointed out already differences in the chemistry of gold from the Witwatersrand and from Barberton. The former contains orders of magnitude more Hg, Zn, Co, As but less Sb. More recently, Barberton gold was found to contain far less Os than Witwatersrand gold which is atypically rich in Os (Frimmel et al. 2005). Hallbauer & Barton (1987) suggested instead a derivation from altered granites in the hinterland, which, in places, contain as much as 80 ppb Au of similar composition. A possible felsic source could be delineated by studying other minerals in the Witwatersrand ore paragenesis that might be typical of such a provenance. One such mineral is uraninite, an important constituent of the auriferous conglomerates, which, in some ore bodies, constitutes an economic U resource.

Uraninite occurs as well rounded to subrounded grains that are typically enclosed, or partially replaced, by pyrobitumen. It is concentrated together with other heavy minerals, such as rounded pyrite, gold, zircon, chromite and many others, along basal degradation surfaces (Feather & Kohn 1975). The spatial association between uraninite and pyrobitumen has been explained by polymerisation and cross-linking of liquid hydrocarbons (Schidlowski 1981). Considering that the bulk of the hydrocarbons formed during diagenesis (England et al. 2002), the rounded uraninite must be older, evidently detrital. This is also supported by highly variable Th/U ratios. High Th contents (up to 4.2 wt%) reported by Feather & Kohn (1975) and Grandstaff (1981) from some uraninite grains are inconsistent with formation from epithermal fluids but point to granitic to pegmatitic sources. Evidence of partial post-depositional U-mobilisation exists in the form of clearly secondary uraninite and other U-bearing minerals (mainly brannerite) as small euhedral grains or minute fills in microfractures.

In this study we analysed both rounded uraninite grains as well as secondary uraninite in microfractures for their trace element concentrations, including Au, with the aim of further elucidating the likely source of the uraninite and possible implications for the source of the gold.

2 Results

Uraninite-bearing pyrobitumen seams from the Vaal Reef at President Steyn mine, the Saaiplaas Reef at Saaiplaas mine and from the B-Reef at the Free State Geduld mine (closed), all in the Welkom goldfield, as well as the B-Reef in the Carletonville goldfield were first studied under an electron microprobe (EMP) for their texture and element distributions. In all cases uraninite grains are embedded within pyrobitumen that forms a stratiform layer on the basal degradation surface

at the bottom of the conglomeratic ore body. The filamentous pyrobitumen also contains rounded pyrite grains as well as visible gold that is interlayered with the near-vertical, columnar pyrobitumen filaments. Back-scatter images and element maps clearly illustrate the presence of two uraninite generations, one being rounded, in places broken up and evidently detrital, the other forming only few micrometres thin, irregularly shaped films along grain boundaries and microcracks (Fig. 1).

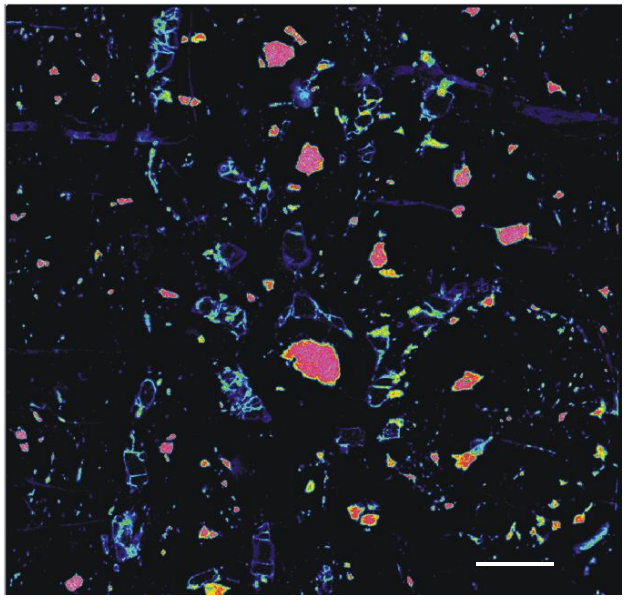

Figure 1. EMP map showing the distribution of U (warm colours indicate high concentrations) in pyrobitumen from the Vaal Reef (President Steyn mine). Note the presence of discrete, subrounded uraninite grains (red) and minute films of secondary uraninite along grain boundaries and microcracks (yellow to green, blue); scale bar = 0.1 mm.

Elements mapped and analysed for by EMP are U, Th, Al, Ti, S, As, Pb, Zn, Fe, Ni, Co, Cr, Bi, Zr, Y, V, Ag, and Au. Galena, sphalerite and arsenopyrite were detected in addition to bitumen, uraninite and gold. Subsequently, the samples were mapped at a high resolution by LA-ICP-MS for the concentrations of As, Y, Mo, La, Ta, W, Bi, Pb, Th, and Au. If individual grains were not large enough for element mapping, spot analyses were obtained.

2.1 Uraninite Chemistry

The uraninite is rich in Pb (≤22.3 wt.%), which is readily explained as in-situ radiogenic product from the decay of U. The Th contents of the rounded to subrounded uraninite are variable, between 1.7 and 4.6 wt.%, thus confirming the detrital nature of these grains. Variable, but generally very small amounts (<1 wt.%) of Fe, Ti, and As were also detected. Of note are elevated Y contents in some uraninite grains (as much as 0.9 wt.%). Similarly to Th, a large variation in Y (some grains are devoid of Y) attests to different sources of individual uraninite grains and thus to their pre-sedimentary age.

Most significantly, the LA-ICP-MS analyses revealed surprisingly high Au concentrations in the evidently detrital uraninite grains. Figure 2 is an example to illustrate this enrichment in Au. It shows the distribution of Pb and Au in two broken-up fragments of a rounded uraninite grain. The (radiogenic) Pb distribution outlines the shape of the uraninite grain. The Au distribution follows almost perfectly that of Pb, clearly indicating that the uraninite is the host of the Au, which reaches concentrations of as much as 12 ppm. Similarly, the two fragments of uraninite show enrichments in Y (≤2.5 wt%), Ce (≤3200 ppm), W (≤100 ppm), and Bi (≤130 ppm). Little within-grain variability in the concentrations of these elements was noted but significant variability exists between grains. Some of the rounded uraninite grains do not contain detectable amounts of Au (<1 ppm) whereas others contain as much as 20 ppm Au. Similarly, Bi and W contents of more than 400 and 550 ppm, respectively, were noted. These relationships indicate that the above elements were original components of the detrital uraninite and not introduced by post-depositional hydrothermal fluids.

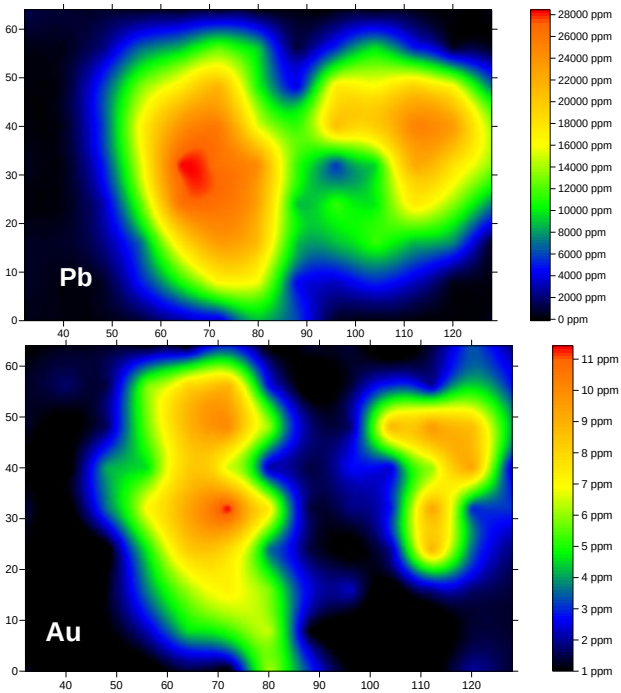

Figure 2. Pb (above) and Au (below) concentration maps of two fragments of uraninite that is embedded in pyrobitumen from the B-Reef (Free State Geduld mine). The Au distribution follows closely that of (radiogenic) Pb and thus the outline of uraninite.

Uraninite that can be considered undoubtedly secondary based on textural grounds was too small to be mapped. Individual spot analyses revealed that this generation has a distinctly different chemistry. It is devoid of detectable Au, As, Bi, and Mo, has very low W and Ta contents and its Y and Th contents are markedly lower than in the larger, rounded to subrounded uraninite grains. For example, rounded to subrounded uraninite grains with Th contents of 3.6 to 4.6 wt.% were found in the same sample together with minute secondary uraninite that contains only 1.2 to 1.8 wt.% Th.

3 Discussion and Conclusions

This study has provided the first trace element data on uraninite from the Witwatersrand goldfields and clearly documents the existence of two generations of uraninite within the auriferous conglomerates. High Th/U in some grains points to derivation from a magmatic or high-temperature hydrothermal source because of the comparatively low solubility of Th in low-temperature hydrothermal systems. These grains correspond to the dominating larger, rounded to subrounded morphological variety and are regarded as detrital. The noted differences in Th, Bi, W, Ta, Mo, Y, Th, As, and Au contents between texturally different uraninite types illustrate that trace element distribution provides a powerful tool to distinguish between detrital and secondary, hydrothermal uraninite, even in cases in which such a distinction is not easily achieved on textural grounds.

A granitic to pegmatitic source of the detrital uraninite is suggested because of the incompatible behaviour of U in magmatic systems, but it is also indicated by the noted relatively high Th contents. A similarly incompatible behaviour of other elemens, such as Y, Ce, Ta, Mo or W, explains the elevated concentrations of these elements in the detrital uraninite grains and their lower concentrations in the secondary, hydrothermal uraninite.

More difficult to explain are the elevated Au concentrations in the detrital uraninite because U and Au are not typically associated with each other. The two elements behave very differently in oxidizing environments, but the observed capacity of uraninite to accommodate significant amounts of Au may reflect formation in a reducing environment.

As not all detrital uraninite grains carry an elevated Au signature, the enrichment in Au must be a feature of the source rock, i.e. some uraninite grains were derived from Au-rich source rocks whereas others were derived from a source with "normal" crustal Au levels.

The pre-Witwatersrand basement consists predominantly of granite-greenstone terranes for which a major crust-forming event at approximately 3.1 Ga in an active, Andean-type continental margin setting is indicated (Frimmel et al., under review). There is evidence of intense late- to post-magmatic alteration of calcalkaline granodioritic to gabbroic rocks, comparable to the alteration around Phanerozoic porphyry Cu deposits (Frimmel et al., under review). As speculated by Hallbauer & Barton (1987) and supported by the unusually high Os contents in the Witwatersrand gold (Kirk et al., 2002, Frimmel et al. 2005), this gold is unlikely to be derived from greenstone-hosted epithermal quartz veins but more likely sourced from granitoids that experienced magmatic-hydrothermal alteration. Mineralization during this kind of late magmatic hydrothermal alteration under reducing conditions might explain the U-Au association discovered in this study. Low-grade but large tonnage disseminated Au (-U) mineralisation in the Mesoarchaean hinterland of the Witwatersrand Basin is thus suggested as potential source for the bulk of the Witwatersrand gold.

Acknowledgements

W.E.L. Minter is thanked for providing some of the analysed samples. Financial support from the German Research Foundation (grant FR 2183/3-2) is gratefully acknowledged. This is a contribution towards IGCP 540.

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