Detrital and hydrothermal origins for quartz, pyrite, and uraninite in Au-U-rich conglomerates of the Witwatersrand gold field, South Africa

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Quartz conglomerate beds in the Witwatersrand gold field, South Africa, ranging from 3.0 Ga to 2.7 Ga in age, host many ore deposits containing gold, uraninite and pyrite. These minerals have been considered by most previous workers as detrital in origin (e.g. Viljoen *et al.*, 1970). This 'model' for the origin of these minerals has been used as important "evidence" for a reduced atmosphere prior to 2.2 Ga because uraninite and pyrite are unstable under oxic conditions (e.g. Holland, 1984). However, some researchers (e.g. Barnicoat *et al.*, 1996) have proposed hydrothermal theories, in which the pyrite and uraninite, as well as gold, are hydrothermal origin. Furthermore, from an examination of the Fe geochemistry of pre- and post-2.2 Ga palaeosols, Ohmoto (1996) suggested that the atmosphere was already oxic 3.0 Ga ago. Because Ohmoto's suggestion contradicts the currently accepted theory for the evolution of atmospheric oxygen, this study was initiated to examine the 'evidence' for detrital origin of uraninite, gold, and pyrite in these deposits.

Occurrences of uraniferous ores

The samples for this study were collected from seven mines in the Witwatersrand Basin: the Western Deep Level South, Western Deep Level East, Free State Saaiplaas Gold, Vaal Reef No.8 Shaft, Joel, Modder B, and Western Holding mines.

The Dominion and Witwatersrand Supergroups overlie the basement comprised of granitoids and greenstones in the Witwatersrand Basin. The Witwatersrand Supergroup mainly consists of fan-, delta-, and shallow marine deposits. They comprise mainly arkosic sediments with detrital minerals of quartz, feldspar, zircon, chromite, sericite, pyrophyllite, illite, and chert. Au-U-rich conglomerate beds occur as thin interbeds throughout the Witwatersrand Supergroup. A highest ore grade bed is only 2–3 mm in thickness and occurs mainly in a basal part of a conglomerate unit above an impermeable shale bed of 0.5–2 m in thickness. The highest ore grade bed is also characterized by the abundance of organic matters with carbon contents of 85–95 wt.%. Diagenetic and/or hydrothermal quartz veins with sulphide minerals often cross cut the Au-U-rich beds.

Textures and compositions of minerals

Quartz grains in the studied samples typically occur as rounded pebbles or granules. Under a cathodeluminescence microscope, quartz overgrowth textures are commonly observed on nearly every grain. The inner part of a quartz grain, which shows bright bluish luminescence, is typically angular to sub-round in shape; the overgrowth and cross cutting quartz show reddish luminescence. The shapes of the inner quartz are similar to those of other detrital minerals, such as zircon and chromite which are slightly rounded but retaining euhedral shapes. Such morphologies of quartz suggest that the inner parts of quartz grains represent detrital grains of quartz, and that the outer parts formed during the diagenetic stage(s) either by groundwater and/or by hydrothermal fluids.

Pyrite also commonly exhibits overgrowth textures, which can be recognized more clearly after etching. Trace element analyses of pyrite grains using an electron micro-probe analyser have revealed that the inner part of a pyrite grain is typically low in trace elements and occasionally contains small amounts of Co. In contrast, the overgrowth zone of pyrite typically contains high amounts of Ni and As, and sometimes Co. These data indicate that the inner and outer parts of pyrite grains were formed at different stages and under different physicochemical conditions. The inner part (first-type pyrite) probably represents detrital pyrite, and the outer part (second-type pyrite) formed during and/or after diagenetic stage(s) (i.e. diagenetic and/or hydrothermal pyrite). The detrital pyrite grains were probably transported together with the detrital grains of quartz and other minerals. However, the shapes of the inner pyrite (detrital pyrite) are typically well rounded, contrasting to the angular to sub-round shales of other detrital minerals in the samples. The second-type pyrite (diagenetic and/or hydrothermal pyrite) is euhedral to subhedral in shape. It often occurs as overgrowths and cross cuts of the secondary quartz. The second-type pyrite also occurs as replacement of chert pebbles while retaining the original texture of chert.

Uraninite and brannerite are closely associated with organic matter. Angular or amoebae shaped uraninite grains are almost always surrounded by organic matter. Uraninite grains that are isolated from organic matter were observed as rounded grains in a sample from the Free State Saaiplaas Gold mine. The rounded uraninite grains occur along grain boundaries of the overgrowth quartz. A halo of reddish luminescence is observed in quartz around the rounded uraninite. Some brannerite crystals are also closely associated with organic matter, but others occur as aggregates of many fine-grained crystals in close association with fine-grained rutile crystals.

Electrum is observed in uraninite, sulphide minerals, and organic matter. Its irregular shape, fracture filling texture, occurrence in veinlets, and other textural features of electrum suggest that electrum is mostly (if not entirely) diagenetic or hydrothermal origin.

Sulphide minerals, other than pyrite, include pyrrhotite, galena, chalcopyrite, gersdorffite, sphalerite and arsenopyrite, all of which appear to be diagenetic or hydrothermal products. The lines of evidence include their occurrences in veinlets; dissemination textures; fracture filling textures; overgrowth textures on grains of pyrite, uranium minerals, and quartz; and irregular shapes.

Pyrophyllite, illite, and sericite are common minerals in high grade ores. These minerals may have been produced from kaolinite by diagenetic processes.

Discussion

The minerals occurring in the Au-U-rich bed can be divided into two types: detrital, and diagenetic and/or hydrothermal. The detrital minerals (and rocks) include: the inner part, which is angular to sub-rounded in shape, of most quartz grains; angular or sub-rounded grains of chert, feldspar, zircon, chromite, and other accessory minerals; and the inner part (round shape) of pyrite grains. Aggregates of rounded uraninite grains, which are extremely rare in occurrence and found to be unassociated with organic matter, also appear to be detrital in origin. The diagenetic and/or hydrothermal minerals include: quartz (overgrowth on detrital quartz); pyrite (overgrowth on detrital pyrite; replacement of cherts; euhedral pyrite crystals); other sulphides (e.g. pyrrhotite, galena, gersdorffite); sericite, illite, and pyrophyllite; most of uraninite and brannerite; and electrum.

It is important to note that the shapes of detrital grains of pyrite and uraninite (rare) are rounded while those of detrital grains of quartz, zircon, and chromite are angular to sub-rounded. The shapes of detrital grains of quartz, zircon, and chromite suggest that the sediments were transported very rapidly and/or short distances from the source regions, probably as storm beds. Therefore, the duration when the detrital grains of pyrite and uraninite was exposed to the air-saturated river water during transportation from the source region to the depositional sites was probably very short, possibly only a few days. The rounded shapes of detrital pyrite and uraninite suggest that the rounding was caused by chemical processes (i.e. dissolution), rather than by mechanical ablation, during transportation and deposition. Dissolution of pyrite and uraninite in short time would have been favoured under an atmosphere rich in both O_2 and CO_2. Rare occurrence of detrital uraninite in the Witwatersrand can also be explained by a model postulating that most detrital uraninite grains were dissolved away in river water during transportation and deposition.

Nearly all the uranium-bearing minerals in the Witwatersrand district are closely associated with organic matter. This close association can be explained by a model proposed by Barnicoat *et al.* (1996) and developed by Ono *et al.* (this volume) for the processes of uranium mineralization in quartz-pebble conglomerate beds of >2.2 Ga. The model is that the formation of uraninite and brannerite in these deposits formed by mixing of two fluids, one was oxygenated groundwater carrying U^{6+}, and the other was petroleum-bearing, reduced fluids. The organic matter in the deposits is suggested to be solidified petroleum. The reduced fluids may also have transported H_2S, As, and heavy metals (Fe, Ni, Cu, Pb, As), and precious metals (Au, Ag) to precipitate diagenetic/hydrothermal sulphides and electrum.

Conclusion

The detrital uraniferous ores in 3.0–2.2 Ga quartz pebble conglomerates contain evidence that the pre-2.2 Ga atmosphere was rich in both O_2 and CO_2.