CRYSTALLOGRAPHIC CONTROLS ON THE WEATHERING OF GOLD

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The majority of studies on the weathering of gold have emphasized geochemical characteristics, postulating the conditions under which it takes place and the degree of dispersion or supergene enrichment that has occurred (Gray *et al*. 1992). Supporting investigations of particulate gold (mostly < 100 µm) have mainly examined the gross morphology and undertaken chemical analyses of external surfaces and, less commonly, of internal sections. For primary (hypogene) grains, observations have included estimation of the amount of pitting and rounding that may have occurred (including quantification of aspect ratios). For secondary (supergene) grains, many of which are pristine (i.e., unweathered) and exhibit subhedral to euhedral crystal forms, there have been some, generally unsuccessful, attempts to determine relationships between crystal morphology and conditions of precipitation. A general conclusion from studies of deeply weathered environments, ranging from tropical rainforests and savannas to warm semi-arid regions, is that the majority of primary gold grains become smaller and increasingly pitted and rounded higher in the profile (Suh & Lehmann 2003, Bowell 1992, Colin *et al.* 1989, Freyssinet *et al*. 1989). These grains commonly contain 5 to > 50% silver; in many instances, there is a thin (1-30 µm), sharply defined, zone of nearly pure gold (< 1% silver) over part or all of the outer margin, in some cases penetrating along cracks into the interior of the grain. This zonation is regarded as being due to depletion of silver, rather than gold precipitation. In comparison, secondary gold grains are commonly composed of high purity gold (999 fineness), although silver-rich grains are reported from environments where thiosulphate ligands may have caused solubilization of gold (Webster & Mann 1984). Some grains of pure 'secondary' gold may be the product of complete depletion of primary grains, but most are considered to have precipitated from groundwater solution. The grains are commonly very small (< 10 µm), in places forming delicate features such as dendrites (Freyssinet *et al.* 1987). Coarser grains (> 150 µm), as aggregates or individual crystals, are rare, though most commonly reported from saline environments such as the southern Yilgarn Craton (Freyssinet & Butt 1988, Lawrance & Griffin 1994).

More detailed investigation of the weathering of gold has been possible through the examination of the external surfaces and internal structures of gold nuggets (that is, masses greater than 4 mm or 1 g). This has also contributed to the debate as to whether nuggets might be of secondary origin. The nuggets are derived from across Australia, from humid tropical Queensland, temperate Victoria and semi-arid Western Australia.

The studies show that all of these nuggets have a primary hypogene origin, but have been modified by secondary processes, i.e., in effect the nuggets are themselves weathering. Features include:

1. Internally, all of the nuggets show a randomly-oriented, polycrystalline microstructure, with many of the crystals displaying coherent twins and/or short incoherent twins that terminate within the crystal. These are typical of thermal annealing at temperatures > 300ºC, implying that the nuggets are primary. There is no evidence for concentric growth fabrics.
2. The nuggets all have appreciable, almost homogeneous, silver contents (> 5% silver) throughout, similar to that of primary gold.
3. There are silver-poor zones deep within nuggets, forming thin (maximum 30-50 µm) veins and as zones along some cracks. These veins and zones have characteristics similar to those of the depletion zones on small grains. The location of silver depletion internal to nuggets is crystallographically controlled, occurring along boundaries between individual crystals, and retains the original crystallographic orientation.
4. Most nuggets, even the largest, have voids penetrating from the surface deep into the interior. The voids may be empty, but the majority contain iron oxides or clays, commonly with fine secondary gold distributed throughout. No secondary minerals are completely enclosed by gold. The voids appear to be initiated along crystal boundaries. Silver depletion on void surfaces, however, is rare.
5. The external surface of some nuggets also have a separate population of mostly anhedral gold crystals of high purity. The crystals are not epitaxial to the host.

Silver depletion is a result of weathering of the gold-silver alloy of the nugget, with the more soluble silver being preferentially dissolved, leaving a pure gold rim to the grain boundary. The process is not one of complete dissolution and reprecipitation, but rather due to the penetration of acid oxidizing fluids along networks of dislocations that provide relative permeability to the alloy, so that only silver is removed. Experiments on worked gold and synthetic alloys have demonstrated that the process is very rapid. The absence of depletion from many external and void surfaces, however, indicates that other mechanisms are operating, involving dissolution of both components of the alloy.

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