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Natural growth of gold dendrites within silica gels

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ABSTRACT

High-grade ores in low-sulfidation epithermal precious metal deposits include banded quartz veins that contain gold dendrites. The processes by which dendrite growth takes place have been subject to debate for decades, especially given that these deposits are known to form from dilute thermal liquids that contain only trace amounts of gold. It is shown here that growth of gold dendrites in epithermal veins at the McLaughlin deposit in California (western USA) originally took place within bands of gel-like noncrystalline silica. The gel provided a framework for the delicate dendrites to form. The high permeability of the gel allowed the diffusion and advection of gold from the thermal liquids flowing across the top of the silica layers to the sites of crystal growth within the gel. Over time, the gel hardened to form opal-A_G. This silica phase is thermodynamically unstable and recrystallized to quartz that has a distinct mosaic texture.

INTRODUCTION

Mineral deposition in low-sulfidation epithermal gold deposits occurs in the shallow subsurface, typically within hundreds of meters below the water table, from thermal liquids that have temperatures of as much as 250 °C. The ore-forming fluids are dilute aqueous solutions with low (<2 mol%) CO_2 contents (Hedenquist et al., 2000; Simmons et al., 2005). The deposits include banded quartz veins, some of which have bonanza-type (>31 g per tonne Au) grades (Hedenquist et al., 2000; Sanematsu et al., 2006; Tharalson et al., 2019; Zeeck et al., 2021). Although the occurrence of gold dendrites having fractal geometries within high-grade banded quartz veins was first described over a century ago (Lindgren, 1915), their mechanism of formation has remained enigmatic (Saunders, 1990, 1994; Saunders and Schoenly, 1995; Sherlock and Lehrman, 1995).

Early researchers, including Lindgren (1915), hypothesized that gold dendrites in low-sulfidation epithermal veins grew within a yielding silica gel deposited along the vein walls, with the gel maturing and recrystallizing to fine-grained quartz over time. This early model was based on hand-specimen observations and the advent of colloid chemistry and crystal synthesis (Holmes, 1917) but has never been tested through microtextural studies on vein samples.

The currently prevailing model links the formation of gold dendrites in epithermal veins to physical processes of metal enrichment (Saunders, 1990, 1994; Saunders and Schoenly, 1995). It advocates that gold colloids are formed deep in the hydrothermal system due to supersaturation, perhaps from boiling. The gold colloids are then mechanically transported by the hydraulic action of the thermal liquid and grow larger through physical aggregation. Ultimately, the nanoparticles are thought to deposit along the vein walls through density accumulation or scavenging from the liquid by charged surfaces on the interface between the vein and the thermal liquid. It is envisaged that upward growth of the dendrites at the tips of the dendrite branches exposed to the thermal liquid occurs simultaneously with the deposition of noncrystalline silica, successively building up a silica layer that hosts gold dendrites. Noncrystalline silica, which forms scales in geothermal power plants (Simmons and Browne, 2000; Reyes et al., 2003; Taksavasu et al., 2018), coprecipitating with the gold thereby provides a framework for the growing gold dendrites and assists in the preservation of the delicate ore textures (Saunders, 1990, 1994; Saunders and Schoenly, 1995).

The two models represent end-member scenarios given that they propose that gold dendrites either form within a previously existing layer of gel-like silica or grow by physical processes of deposition in open space. We provide textural observations suggesting that gold dendrites can indeed grow within layers of silica gel, as originally envisaged more than a century ago, which does not necessitate gold transport in colloidal form in the thermal liquids. The textural evidence presented was discovered in high-grade ores from the McLaughlin deposit in California (western USA), which represents a young (<2.2 Ma; Sherlock et al., 1995) and well-preserved low-sulfidation epithermal deposit that has not been subjected to tectonic or metamorphic overprint.

GEOLOGICAL SETTING

The McLaughlin deposit is located ∼120 km north of San Francisco within the Coast Ranges of California. Mineralization is located in the structural footwall of the Stony Creek fault separating the Middle Jurassic Coast Range ophiolite in the southwest from Upper Jurassic sedimentary rocks of the Great Valley sequence to the northeast (Sherlock et al., 1995; Tosdal et al., 1996). The deposit was mined by open pit between 1983 and 1996 CE. It contained a total pre-mining mineral resource of 24.3 million tonnes of ore grading 4.49 g per tonne gold (Tosdal et al., 1996). The main ore body was a pipe-like sheeted vein complex that developed within a dilatant zone between tholeiitic basalt and a mélange consisting of sedimentary rocks and serpentinite. The sheeted vein complex is a zone, as much as 100 m in width, that is composed of crosscutting veins that are centimeters to meters in width (Sherlock and Lehrman, 1995; Sherlock et al., 1995; Tosdal et al., 1996).

METHODS

Field work at the McLaughlin deposit included representative sampling of high-grade ores (Sherlock et al., 1995). Opaline vein samples used for this study were taken from the 1580

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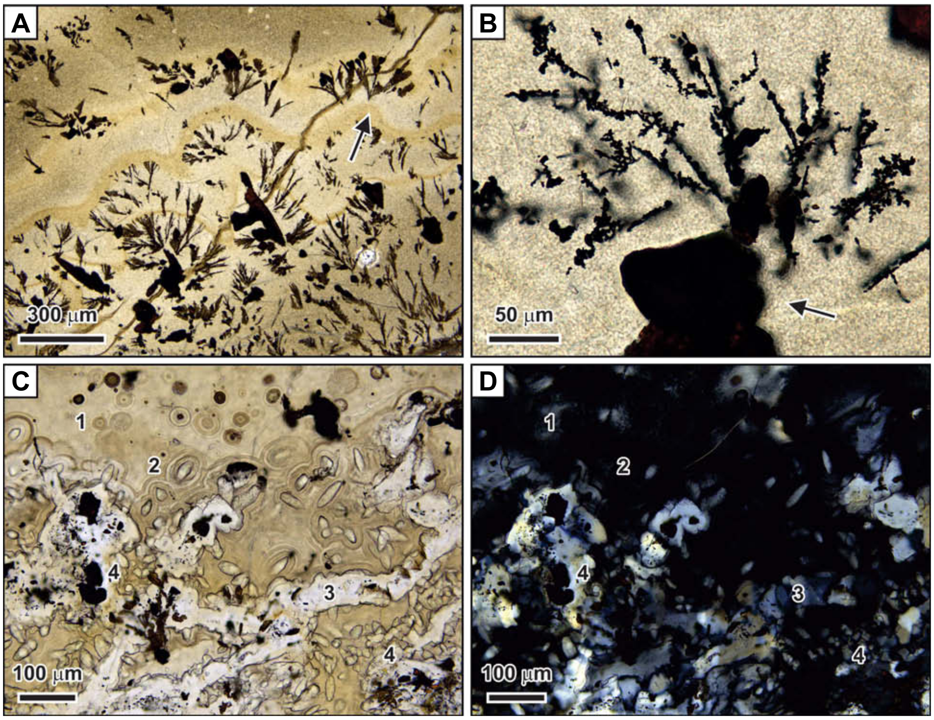

Figure 1. Microtextures in opaline veins from the McLaughlin deposit in California (western USA) observed in thin sections in plane-polarized (A–C) and crossed-polarized light (D). (A) Gold and sphalerite dendrites within an opal-A_G matrix. Opal-A_G band hosting dendrites has a wavy upper surface indicative of hydraulic shaping (arrow). The large opaque crystals are pyrargyrite. (B) Gold dendrite grown within an opal-A_G matrix. The silica matrix surrounding the large pyrargyrite crystal appears to be slightly deformed (arrow). (C) Recrystallization textures developed in silica matrix. Concentrically banded silica spheres having a heterogeneous turbid appearance occur in the opal-A_G matrix (1). Doubly terminated quartz crystals are present within cores of the concentric features (2). Zones of completely recrystallized quartz are present (3). Some of the gold and sphalerite dendrites, as well as the pyrargyrite crystals, are surrounded or encapsulated by quartz (4). (D) Image of the same field of view as in C in crossed-polarized light showing that the matrix containing relict microsphere is at least partially isotropic. The mosaic texture is caused by intergrowth of anhedral quartz grains having interpenetrating grain boundaries.

bench (feet above sea level) of the sheeted vein complex. Polished thin sections were obtained and studied petrographically. Semiquantitative chemical analyses of ore minerals were performed on a TESCAN MIRA3 LMH Schottky field emission–scanning electron microscope equipped with a single-crystal yttrium aluminum garnet (YAG) backscatter electron detector and a Bruker XFlash 6-30 silicon drift detector for energy-dispersive X-ray spectroscopy. The instrument was operated at 15 kV using a working distance of 10 mm.

TEXTURAL RELATIONSHIPS

The opaline veins at the McLaughlin deposit are composed primarily of relict silica microspheres, which are mostly isotropic under crossed-polarized light. The relict microspheres of opal-A_G (cf. Smith, 1998), which are presumably deformed and slightly modified through recrystallization, have spherical to oblate shapes, are 1–5 μm in size, and are variably fused together. Due to the spherical shapes of the relict microspheres, cavities between them have sickle-like shapes. Adjacent bands of silica show subtle differences in color and packing density. Individual bands of opal-A_G are of variable thicknesses, commonly exhibiting botryoidal and wavy surfaces (Fig. 1A).

The veins contain many bands of opal-A_G that are host to ore minerals. The ore minerals occur as single crystals or polycrystalline aggregates within the silica matrix. Most notable are dendritic aggregates of gold (millesimal fineness of 593–790, $n = 70$). The gold dendrites form as aggregates as much as 500 μm long which are intergrown with sphalerite and minor pyrargyrite (Figs. 1A and 1B). Large dendritic gold aggregates are oriented approximately perpendicular to the silica bands, whereas smaller dendrites are randomly oriented (Figs. 1A and 1B). Mineralized bands also contain delicate sphalerite dendrites as well as euhedral to subhedral crystals of pyrargyrite that are as much as ~200 μm in size. In some cases, the opal-A_G matrix varies in packing density around the larger stubby pyrargyrite crystal (Fig. 1B), possibly suggesting that the matrix was yielding during crystal growth and pushed aside by the growing crystals. Minor pyrite crystals ranging as large as ~100 μm are present, some of which contain galena inclusions.

The opal-A_G in the mineralized bands is variably recrystallized, exhibiting a range of textures. This includes concentrically banded silica spheres having a heterogeneous turbid appearance (Fig. 1C). In plane-polarized light, the <50-μm-sized silica spheres differ in color from the surrounding opal-A_G matrix, and some may have matured to a degree where small doubly terminated quartz crystals have nucleated in the cores. When near to one another, these evolved to complex quartz aggregates of doubly terminated quartz crystals that are suspended in the matrix (Fig. 1C). In some locations, growth of the quartz crystals resulted in the formation of massive clear quartz (Fig. 1C). In crossed-polarized light, these zones exhibit a mosaic texture in which quartz grains have irregular and interpenetrating grain boundaries and differ in orientation (Fig. 1D). Relict microspheres are common in the mosaic quartz and can be identified by high-magnification optical microscopy. In many cases, clear quartz aggregates and crystals formed through recrystallization of the opal-A_G encapsulating ore mineral dendrites or small complexly shaped aggregates of ore minerals (Figs. 1C and 1D).

MECHANISM FOR DENDRITE GROWTH

The textural relationships at the McLaughlin deposit suggest that the gold dendrites have grown within the layers of noncrystalline microspherical silica, not solely at the interface between the silica host and thermal liquid. The delicate dendrites occur throughout the opal-A_G layers and not preferentially at the top of the bands. They are commonly oriented perpendicular to the bands and appear to have grown toward the top of the microspherical matrix that hosts them based on the shape of the dendrite branching. Although some of the larger gold aggregates are oriented, many of the small dendrites are randomly oriented, arguing against a model of dendrite growth taking place only along tips exposed in the open part of the vein. The silica provided a framework in which the delicate dendrites grew.

The gold hosted within individual colloform bands of noncrystalline silica at McLaughlin may have formed by a mechanism of crystal growth not unlike that of the synthesis of crystals in silica gels under laboratory conditions (Brenner et al., 1966; Kotru et al., 1986; Oaki and Imai, 2003; Raj et al., 2008). In crystal synthesis, a silica gel is obtained through slight acidification of sodium metasilicate (water glass) following impregnation with a reactant. An overhead feed solution is added. Within days, downward diffusion of the feed solution causes the growth of crystals in the chemically inert gel which provides a three-dimensional

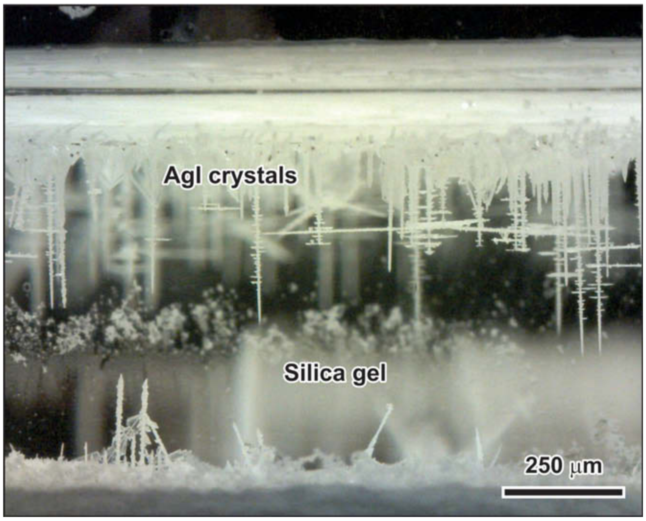

Figure 2. Delicate silver iodide (AgI) crystals experimentally grown in silica gel. The gel was prepared from 20 ml of sodium metasilicate solution and 10 ml of 1 N acetic acid. It was then mixed with 10 ml of 3 M $AgNO_3$ and allowed to mature at 25 °C for one week. After maturation, 10 ml of 3 M potassium iodide (KI) solution was poured over the top of the gel. The chemical reaction of $AgNO_3$ and KI initially started at a slow rate to precipitate small white to colorless AgI crystals near the boundary between the gel and KI solution. Over the next seven days, the reaction front migrated down in the beaker, and the gel behind the reaction front became clear and large AgI crystals grew within the clear silica gel. Experimental setup is from Brenner et al. (1966) and Halberstadt (1967).

structure in which the crystals grow and are held in position of their formation. Different crystal morphologies develop as a function of the degree of supersaturation, with polyhedral forms developing at near-equilibrium conditions and dendritic shapes being dominant at far-from-equilibrium conditions (Oaki and Imai, 2003).

To illustrate that textures similar to those observed at McLaughlin can be synthesized, we conducted a simple laboratory experiment growing delicate silver iodide dendrites in a gel (Fig. 2). Similar to this crystal growth experiment, gold at McLaughlin may have formed through deposition of a silica gel along the vein walls, followed by growth of gold dendrites within the gel layer by diffusion or advection of solutes through the gel from thermal liquids flowing across the top of the silica layer. However, the experimental growth of silver iodide does not replicate the observation that most of the dendrites in natural samples grew upward toward the top of the gel layers, perhaps because the experiment was not designed to mimic flow conditions in veins.

Quantitatively, the high abundance of gold in the opal-A_G layers at McLaughlin suggests that the silica and the gold were not coprecipitated from the thermal liquids. At 230 °C and a salinity of 2.5 wt% NaCl (Sherlock et al., 1995), 1 L of thermal liquid has a silica solubility of 0.278 g L^{-1} at 27.6 bar (cf. Akinfiev and Diamond, 2009), which is equivalent to 0.133 cm^3 of opal-A_G. At a content of 2 ppb Au (Simmons et al., 2016), 1 L of thermal liquid contains 0.556×10^{-9} g Au and could form a grain of gold that is 0.288×10^{-10} cm^3 in size. On the scale of Figure 1B, this would mean that a cube of opal-A_G having a dimension of $100 \times 100 \times 100$ μm would contain a gold grain that is $0.06 \times 0.06 \times 0.06$ μm in size. The observed abundance of gold is clearly much higher (Fig. 1B). If the silica gel formed from silica colloids transported by the thermal fluids (Saunders, 1990), only a small percentage of the silica would be deposited to form the gel layer, presumably because silica colloids have a negative surface charge (Abendroth, 1970) limiting aggregation and deposition. In contrast, gold formed through metal transport to the sites of dendrite growth by a large amount of liquid flowing over the top of the layers of previously accumulated silica gel. The difference in depositional mode and growth mechanism between silica and gold are envisaged to be responsible for the significant precious metal enrichment.

RECRYSTALLIZATION

Following growth of the gold dendrites in a yielding gel matrix, the gel must have hardened to form opal-A_G, which is now making up the mineralized bands in the veins from the McLaughlin deposit. However, the microtextural evidence suggests that the opal-A_G continued to mature and recrystallize given that some of the originally microspherical bands are now composed of mosaic quartz (Figs. 1C and 1D).

Investigations on silica deposits formed by hot springs (Herdianita et al., 2000; Lynne and Campbell, 2004; Rodgers et al., 2004; Lynne et al., 2005) and silica scales in geothermal power plants (Reyes et al., 2003; Raymond et al., 2005) confirmed that opal-A_G is thermodynamically unstable. Maturation involves the transformation of noncrystalline opal-A_G into opal-CT, which in turn recrystallizes into opal-C and then into quartz. Maturation of noncrystalline silica to quartz has been achieved in hydrothermal experiments in as little as days to months (Bettermann and Liebau, 1975; Oehler, 1976). Textural reequilibration is heterogeneous in the vein samples investigated, with some colloform bands still being at least partially isotropic in crossed-polarized light while others are largely recrystallized. Complete textural reequilibration results in the formation of a quartz mosaic texture, which is the most common quartz texture encountered in epithermal deposits worldwide (Dong et al., 1995; Moncada et al., 2012).

IMPLICATIONS

The textural observations made at the McLaughlin deposit suggest that gold dendrites contained in low-sulfidation epithermal veins can form through growth within silica gels. In contrast to the model proposed by Saunders (1990), gold dendrite growth within silica gels is not dependent on colloidal metal transport in the thermal liquids, which may explain why aggregates of gold and sphalerite have similar morphologies in the investigated samples. The textural evidence is also not consistent with earlier studies at McLaughlin highlighting the importance of hydrocarbons in scavenging colloidal gold from the thermal liquids by hydrostatic attraction (Sherlock et al., 1995).

Growth of the ore metal dendrites in silica gels at far-from-equilibrium conditions has implications for the understanding of the hydrothermal processes occurring in the epithermal environment. Initial deposition of the noncrystalline silica requires the thermal liquids to periodically reach extreme supersaturation (Saunders, 1990; Simmons and Browne, 2000). Such high degrees of silica supersaturation are most likely achieved through vapor loss associated with catastrophic pressure drops accompanying hydrothermal eruptions on the surface. It is envisaged here that the initial rapid deposition of the gel-like silica layers along faults during such events of fluid flashing was followed by more sustained periods of nonviolent two-phase liquid plus vapor flow. Metal-laden thermal liquids flowing through the open spaces along the structures controlling vein formation provided the source of gold and other elements for diffusion or advection of elements through the previously formed silica gel, enabling growth of the delicate dendrites. Repetition of this process through time results in the development of banded epithermal veins containing multiple layers of high gold grades that alternate with barren quartz bands that record periods of more quiescent fluid flow.

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