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HYDROTHERMAL SILICA – GOLD STALACTITES FORMED BY COLLOIDAL DEPOSITION IN THE CIROTAN EPITHERMAL DEPOSIT, INDONESIA

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ABSTRACT

Hydrothermal stalactites have been discovered in the deepest level of the Cirotan mine (Western Java), an adularia–sericite type of precious metal epithermal deposit of Pliocene age. These stalactites show a succession of colloidal silica and gold-rich sulfide bands. Oxygen isotope geochemistry indicates that hydrothermal fluids were of meteoric origin, similar to present rain water in the area. Quartz textures indicate early deposition by gel related to silica supersaturation, then of alternating bands related to deposition and drying, as shown by desiccation marks. Stalactite formation and the origin of alternating bands could be related to local input of magmatic gas.

Keywords: hydrothermal ore deposit, gold–silver, stalactites, silica gel, Indonesia.

SOMMAIRE

Des stalactites hydrothermaux ont été découverts dans les niveaux les plus profonds de la mine de Cirotan, un gisement épithermal de type adulaire–séricite à métaux précieux d'âge Pliocène de l'Ouest de Java. Ces stalactites montrent une succession de dépôts de silice d'origine colloïdale, et de sulfures aurifères. La géochimie isotopique de l'oxygène du quartz montre que les fluides étaient d'origine météorique et similaires aux pluies actuelles. Les textures du quartz indiquent un premier dépôt par forte saturation des fluides en silice, provoquant des dépôts gélatineux, puis des alternances de dépôt et d'assèchement, marqués par des traces de dessication. La formation des stalactites, et l'origine des alternances, pourraient être en relation avec une arrivée locale de gaz magmatique.

Mots-clés: gisement hydrothermal, or, argent, stalactite, gel de silice, Indonésie.

INTRODUCTION

Mineral deposition in hydrothermal systems related to subaerial volcanism has been extensively studied, especially in modern environments, such as those found in the North American Cordillera, New Zealand and Japan (Elder 1981, Fournier 1985, Hedenquist 1992). The hydrology of these geothermal systems, however, remains poorly known at depth (>500 m). Speleothems, like stalagmites and stalactites, are classical indicators of hydrological processes in cave environments. Exceptionally preserved *in situ* stalactites and stalagmites were discovered in 1992 in the Pliocene epithermal gold deposit of Cirotan, West Java. In this paper, we present the results of a detailed mineralogical and geochemical study of these textures and discuss their origin.

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GEOLOGICAL CONTEXT

Sumatra, Java and most Indonesian islands form part of the western Sunda–Banda continental arc margin, which was developed along the northern margin of the subducting Indian–Australian plate during Cenozoic collision with the Eurasian plate (Carlisle & Mitchell 1994). Cirotan, the largest mine of the Cikotok district of West Java, produced more than 5 t Au and 140 t Ag until its closure in 1994 (Fig. 1). The deposit consists of a large vein hosted by a Pliocene microdiorite (4.5 Ma) that cross-cuts a Miocene volcano-sedimentary sequence (9.5 Ma). K–Ar dating on vein minerals gave an age of

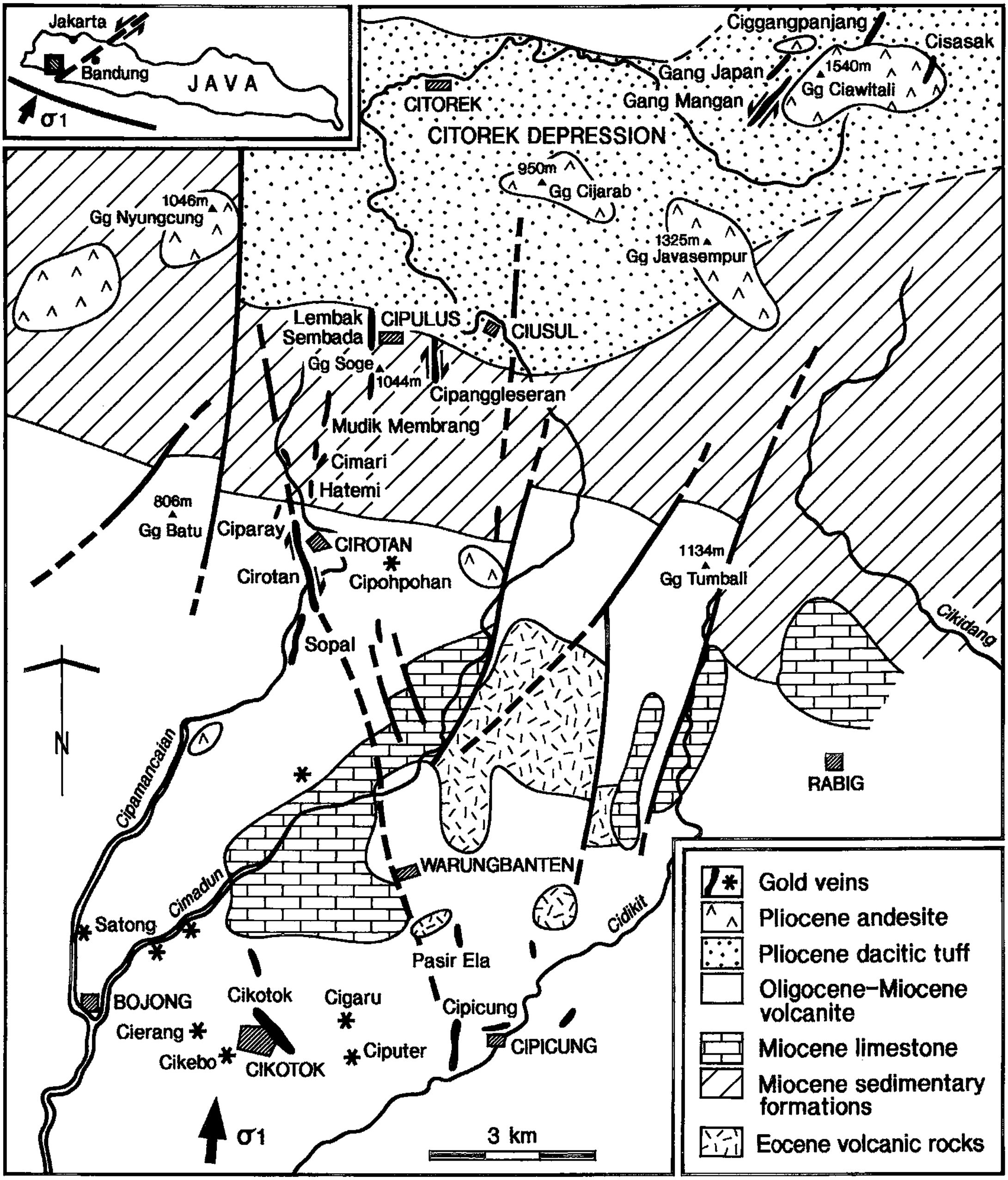


FIG. 1. Location of the Cirotan mine in the Western Java gold district.

1.7 ± 0.1 Ma, which corresponds to the Pliocene–Pleistocene boundary (Milési *et al.* 1994). Vein formation resulted from a single mineralizing event, with five stages of deposition characterized by: (I) siliceous replacement of host rock, (II) thick siliceous breccias containing mainly pyrite, (III) thick cockade breccias with polymetallic sulfide ore and local concentrations of rhodochrosite and rhodonite, (IV) late precious-metal-bearing brecciated bonanzas in lenticular bodies, enriched in W, Sn, Au and Bi, and (V) barren vuggy quartz (Marcoux *et al.* 1993, Milési *et al.* 1994). Intense argillic alteration of the hanging wall is superimposed on a regional propylitization. The mineralogy of the deposit is thus mostly characteristic of the adularia–sericite (low-sulfidation) epithermal type of Hayba *et al.* (1985), with some affinities toward W–Sn deposits. Cirotan is therefore interpreted as a hybrid deposit transitional between the adularia–sericite epithermal type and the porphyry-tin deposit type (Milési *et al.* 1994).

Studies were carried out on quartz- and sphalerite-hosted fluid inclusions from seven samples representatives of the five stages of deposition (Nehlig & Marcoux 1992). The fluids are characterized by moderate salinities (2.89 to 7.15 wt% eq. NaCl) and show a progressive increase in salinity from the initial stage of silicification until the bonanza stage (IV). The fluid homogenizes in the liquid phase between 207° and 292°C. If one assumes an unconstrained hydrostatic column to the surface and almost no CO_2 content, the absence of phase separation within fluid inclusions is consistent with erosion of a minimum of 410 m since mineralization or recrystallization (Barton & Chou 1993). Sulfur isotope compositions of pyrite and galena (average $\delta^{34}S$: 2.2 and 1.0‰ respectively) are close to the 0‰ isotopic value typical of mantle-derived magmatic sulfur. These data, combined with the late enrichment in W, Sn, Bi and Au, reflect an increasing participation of a magmatic component during the mineralization process (Milési *et al.* 1994). The Cirotan deposit is thus considered to reflect a genetic link with porphyry-tin deposition and could be explained by a progressive upward migration of an underlying magma (Milési *et al.* 1994), or upward migration of its thermal aureole above a stationary pluton.

MINERALOGY AND STRUCTURE OF CIROTAN SPELEOTHEMS

Meter-size elongate cavities filled with either quartz–sulfide stalactites or flowstones appear at the bottom of the mine workings (330 m level), on the hanging wall of the Cirotan vein (Fig. 2). Stalactites are up to 15 cm long and 8 cm in diameter (Fig. 3). Stalactites display mostly cylindrical shapes, locally bulbous and with pendant forms hanging from the edges of irregular vein-wall surfaces. Isolated stalactites display a perfectly circular section and rounded tip. Their axes are vertical, indicating that deposition was influenced by gravity and that no tilting occurred since their formation. Centimeter-thick flowstones formed by films of water flowing downward on the walls of the cavity (Hill & Forti 1986) seem to be in continuity with the stalactites and display a similar mineralogy (Genna *et al.* 1996). At the bottom of the cavity, stalagmites are poorly developed, but may reach a few centimeters in height. The bottom also is partly filled with detrital quartz, iron oxides and cemented angular fragments of collapsed stalactites. A micropaleontological study did not result in evidence of biological remnants.

Whole-rock chemical analysis of a stalactite using X-ray-fluorescence (XRF) and inductively coupled plasma with mass spectrometry (ICP–MS) methods indicates a strong enrichment in Au (1.5 ppm), Ag (15 ppm), W (0.1%), As (294 ppm) and Zn (3.43%). Sulfides are abundant. A detailed paragenetic study across several stalactites revealed the same sequence of deposition of colloform microcrystalline silica mineral and sulfides in each case (Fig. 3). The paragenetic sequence of stalactites was divided into four mineralogically distinct layers, numbered outward from the center of the stalactite (Fig. 3).

Layer 1 is homogeneous and composed of skeletal pyrite and quartz, nucleated on early quartz grains, with evidence of recrystallization resulting in plumose-textured, microcrystalline, length-fast quartz (Sander & Black 1988, Garlick & Jones 1990, Flörke *et al.* 1991). Skeletal pyrite is invariably formed as a pseudomorph after acicular pyrrhotite (Fig. 3d). Despite its filamentous shape, the discontinuity of the pyrite network seems to preclude a bacterial origin, as found in some iron–silica marine deposits (Juniper & Fouquet 1988). A second type of pyrite, which appears as small primary euhedral crystals, locally encrusts the skeletal pyrite. This second type becomes abundant at the boundary of layer 1, forming porous aggregates associated with scarce patches of galena and sphalerite. This layer seems to correspond to the recrystallization of an initial mass of gel.

Layer 2 is composed dominantly of light-brown sphalerite with minor galena, rare chalcopyrite and quartz; sphalerite does not display evidence of the "chalcopyrite disease" texture. **Layers 3 and 4** contain tiny microspheres composed mainly of recrystallized to microgranular grains of quartz a few micrometers in diameter (Fig. 4), larger in layer 4 than in layer 3. Interpenetrating grain-boundaries result in a jigsaw texture, and are interpreted as the product of crystallization from an amorphous state, which corresponds to a dehydration process (Lovering 1972). At the scanning electron microscope scale, these spherules contain a nucleus of a detrital mineral, coated by microcrystalline quartz and, in some cases, rhodonite and sulfide. These layers are much poorer in sulfides,

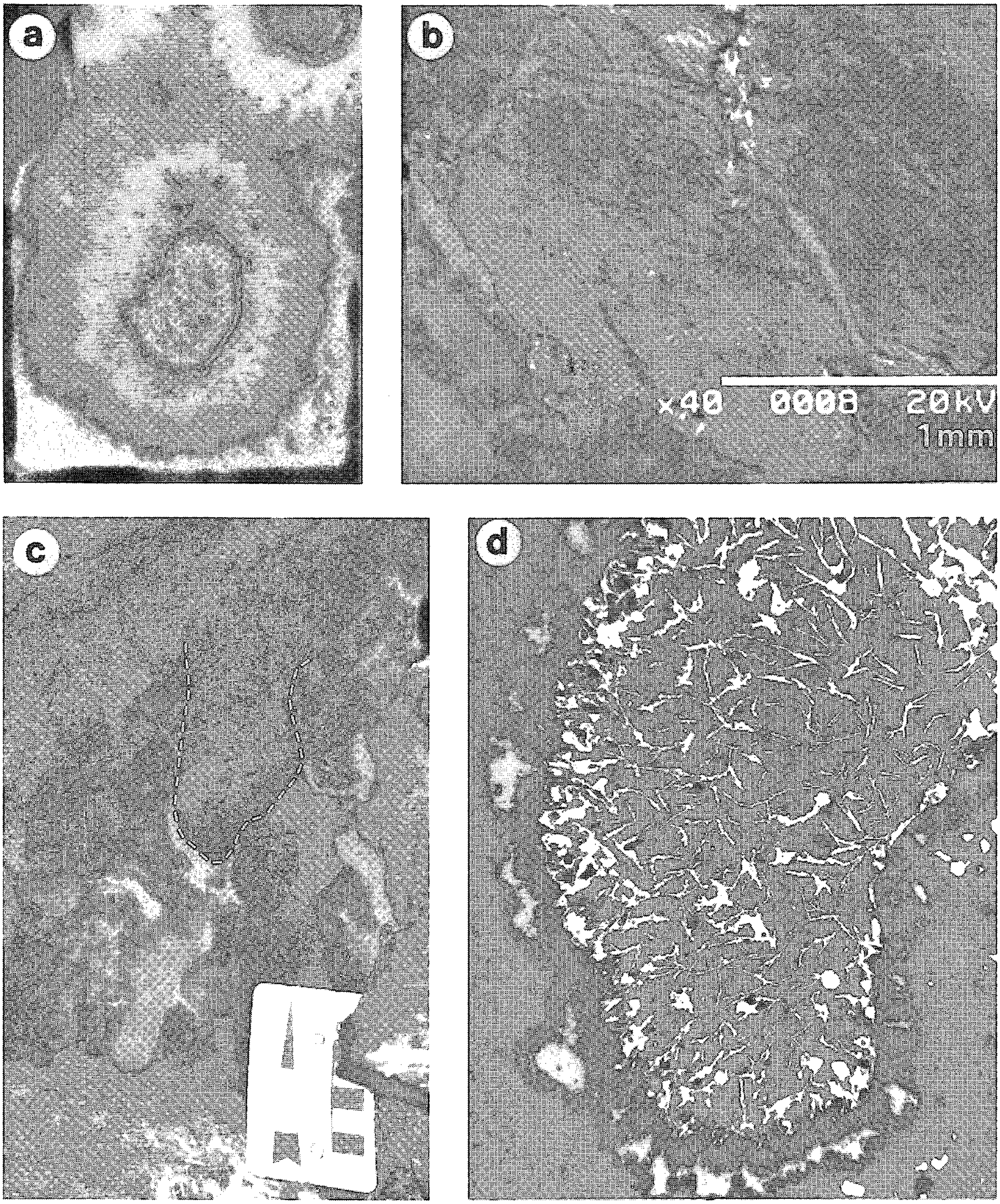
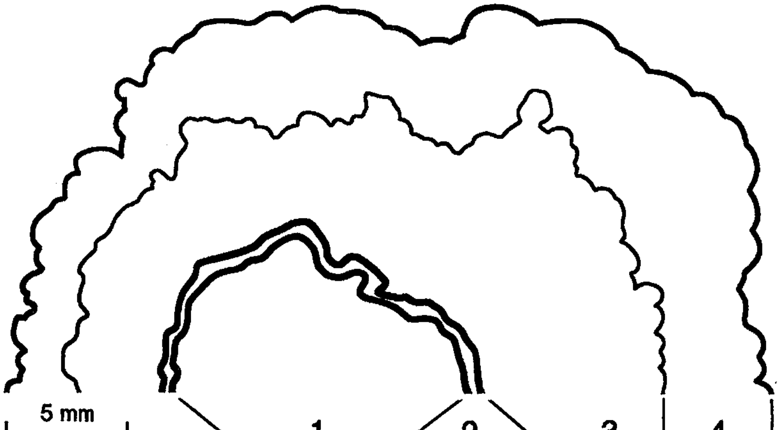


FIG. 2. a. Cross-section of a stalactite (actual size: 3 cm across). b. Stalactite desiccation textures observed on the surface of a band. c. Cavity showing the stalactites on the hanging wall of the Cirotan vein; the dashed line outlines the largest stalactite (scale in cm). d. Detail of the first layer, showing pseudomorph of pyrrhotite crystals by pyrite in a matrix of quartz. Width of field of view: 1 cm.

represented by isolated grains of galena, chalcopyrite and primary euhedral pyrite. The latest generation of microcrystalline quartz shows mm-size terminations pointing toward open cavities.

Laser-ablation microprobe – inductively coupled plasma – mass spectrometry (LAM–ICP–MS) analyses performed across the layers do not indicate any evidence of chemical evolution from the core to the



	1	2	3	4
Quartz	massive	—	banded	rosette
Remnant texture	rosette			
Recrystallization	high	—	—	high
Sulfides	pyrite pyrrhotite	galena sphalerite chalcopyrite	pyrite	

FIG. 3. Detailed cross-section of a stalactite, and main features of layers 1 to 4. Diameter of the stalactite is 2 cm.

periphery of the layers. Pyrite is chemically similar in the different layers: we found small amounts of Co (16 to 20 ppm) and As (10 to 40 ppm) and an almost constant Ni/Co value (1.8 to 2). Gold and tungsten were not detected at that scale, but are probably associated also with pyrite. Mineralogical features do not yield unequivocal evidence concerning the timing of stalactite formation in the paragenesis, but open-space crystallization and high values of W (0.1%) in stalactite material are typical of the end of the depositional process. The Cirotan speleothems were therefore formed either late in stage 3 or, more probably, during stage-4 mineralization. Paleobarometric reconstruction, using data on fluid inclusions in minerals from the same level as the speleothems, indicates a minimum depth of deposition of 790 m (Nehlig & Marcoux 1992).

The surfaces of the stalactite layers were studied by selective dissolution of silica using dilute HF. They display a botryoidal texture, which is generally regarded as evidence of a pre-existing colloid (Krauskopf 1979). Fractured surfaces on aggregates of microspheres locally show a polygonal network of ridges that may correspond to either anhedral boundaries of quartz grains or desiccation features. The same kind of marks has been found on rhodonite layers, which suggest that it corresponds to a desiccation process after mineral deposition.

OXYGEN ISOTOPE GEOCHEMISTRY

The oxygen isotopic geochemistry of 36 quartz samples from the Cirotan deposit was studied. The methodology and results are summarized on Figure 4. The values for $\delta^{18}O$ vary between + 5.1 and + 8.1‰. These values are fairly homogeneous. Stalactites display the same isotopic signature as the other forms of quartz. Computation of the $\delta^{18}O$ value of water in equilibrium during deposition was performed following Matsuhisa *et al.* (1979). Recrystallization of the colloidal materials precludes any confidence in fluid-inclusion data from the stalactites. Therefore, a temperature of 250°C was used, as indicated by fluid-inclusion data from the deposit (Nehlig & Marcoux 1992) and studies of similar deposits (Hayba *et al.* 1985, Heald *et al.* 1987). The resulting values range between –4.4 and –1.6‰, and are indicative of meteoric water (Fields & Fifarek 1985). They seem

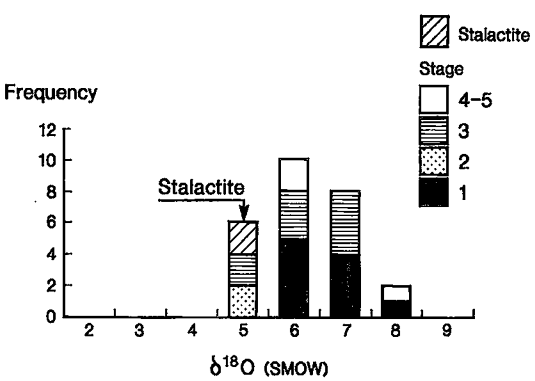


FIG. 4. Histogram of oxygen isotope results in quartz of the Cirotan mine; 1 – 5: stages of ore formation.

very similar to the present $\delta^{18}O$ values of rain in Java, which averages –4‰ (International Atomic Energy Agency 1969–1979).

DISCUSSION

Origin of the stalactites

Hydrothermal stalactites are rare features. Cunningham *et al.* (1994), Plumlee (1994) and Campbell & Barton (1996) have recently described structures in Porco (Bolivia), Creede (Colorado), and in several other epithermal deposits. Two types of growth have been identified: (1) the soda-straw type is formed as a downward-growing collar around an orifice through which water flows. Precipitation would correspond to the formation of classical speleothems. This type of stalactite records the presence of an interface between liquid water and either atmospheric or hydrothermal vapor. (2) The dripping type (icicle type) is related to deposition along the margin of a pendant drop; these stalactites tend to be conical. This dripping process may be caused by evaporation of an aqueous fluid, leaving a nonvolatile residuum, or by mechanical flow, with distortion of a plastic gel, as has been proposed by Shaub (1955) and Landmesser (1988) in order to explain some of the features observed in agate. This latter process could occur in either a liquid or gaseous medium and, therefore, does not have the same hydrological implications as the soda-straw type.

The stalactites at Cirotan differ in several ways from the soda-straw type described at Creede: (i) they do not display any soda-straw features; (ii) they are polymineralic, with simple but recurrent stages of mineral deposition; (iii) they reveal chemical characteristics close to those of bonanza ores; (iv) they do not show evidence of successive episodes of dissolution. They belong, therefore, to the dripping type. However, their morphology is not conical, and a formation of the early stage (layer 1) of the stalactite might therefore be related to a different process. Taking into account the colloidal origin of the silica in the core of the stalactite, the Cirotan speleothems could have been formed initially from the dripping of a viscous gel in a hydrothermal solution. In order to form such a gel, amorphous silica must be mobilized and accumulated to concentrations well in excess of the quartz-saturation level. If the degree of supersaturation becomes relatively high by temperature decrease or ligand complexing, monomeric silica polymerizes to chain and cyclic species, and gelification occurs throughout the solution. Such a process could result from a sudden reduction in pressure and temperature of the solution, whereby the fluid became highly viscous, and gelatinous masses accumulated along the flow path (Flörke *et al.* 1982, Weres *et al.* 1982, Saunders 1990). However, the detailed mechanism for such a process involving viscous dripping needs to be more quantitatively assessed.

After the formation of the core of the stalactite, deposition is controlled by pre-existing solid surfaces and could be related to surface evaporation (layers 2 to 4). This suggests that the rate of gel deposition could be lower. Such a process occurs if the supersaturation of the solution remains relatively low, with a dominant polymerization of the amorphous silica. Depositional features indicate that the hydrothermal conduits were largely unfilled at that time. In such a medium, gel deposition could take place directly on the surface of the stalactite through interaction with an amorphous-silica-supersaturated fluid, or by the downward movement of a surficial film. The local presence of branching of the stalactite suggests that the former process was the more important.

The formation of stalactites at Cirotan suggests that an evolution of the hydrothermal medium occurred, with the degree of silica supersaturation decreasing from the beginning to the end of the depositional process.

Gas–liquid interface

The presence of cracks between layers in the stalactites at Cirotan shows that several episodes of desiccation probably occurred in the silica gel. Gel desiccation could occur in the presence of either a gas or a liquid, but the desiccation cracks have been observed on the surface of layers 3 and 4, which are related to surface evaporation, and which could only have occurred in a gaseous medium. Industrial experiments on siliceous gel formation demonstrate that, for low temperatures (25–45°C), gelification appears in a few hours to a few weeks in a non-agitated medium (Oehler 1976). This succession of desiccation and deposition allowed the integrity of the banding to be preserved (Heaney 1993). The process of supply of liquid was therefore intermittent, and deposition at Cirotan is inferred to have occurred in a series of pulses, with variation from gaseous to liquid media, with enough time to dry the gel between each stage of deposition. After construction of the core, stalactite formation involved flow of hydrothermal solution, gel formation, coating of the wall of the cavity, and dripping.

The drying process is indicative of a transition from liquid to gas. In epithermal deposits of the adularia–sericite type, two zones of transition may be distinguished. At the bottom of the hydrothermal system, rapid decompression of a high-temperature fluid caused boiling and fluid immiscibility (White *et al.* 1971). Fluids could be expected to travel at different rates in the fractured media and lead to alternate inputs of gas and liquid. This situation would correspond to a high-vapor-pressure zone. At the top of the hydrothermal system, the fluid could also be in contact with the atmosphere or with a low-vapor-pressure zone (Goff & Gardner 1994). Variations in the level of the water table can be expected to occur as in volcanoes, caused either by pulsative degassing, doming, or caldera collapse (Cortini & Barton 1993, Elston 1994, Sillitoe 1994); these variations of the altitude of the water table could be also responsible for coating and drying the speleothems at Cirotan. However, no remnant was found that could allow us to determine the precise nature of the gas that dried the gel during stalactite formation. The association of stalactite formation with the late hydrothermal stage and enrichment of the stalactites in tungsten suggest a magmatic relation with the bottom of the system. More work is needed for the characterization of these two zones of gas–liquid transition in present geothermal system, in order to get a better understanding of the significance of stalactites in epithermal vein-type deposits.

CONCLUSIONS

An adularia–sericite type epithermal deposit generally is considered to be the result of the cooling of hydrothermal water, and the nonmagmatic component usually dominates. The Cirotan deposit, in western Java, displays a similar origin, as shown by the isotopic signature in the oxygen of the quartz, identical to that of the present rain-water.

Stalactites discovered in the deepest level of the Cirotan mine show a succession of colloidal silica and gold-rich sulfide bands. Quartz textures reveal an early deposition of gel related to silica supersaturation, then alternating episodes of deposition and drying, as indicated by desiccation marks. Stalactites are therefore the expression of the variable position of the gas–liquid interface, indicating either large variation in the position of the water table or, more probably, an episodic input of magmatic gas.

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