Speleothems and spiders: morphology and origin of gypsum nucleated on spider webs, Deer Cave, Sarawak, Borneo

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Abstract: An unusual form of gypsum is reported from Deer Cave, Gunung Mulu National Park, Sarawak, Malaysia. The gypsum is originally derived from decomposing bat guano, is air-dispersed, dissolved in condensation water entrained in spiders’ webs, and then recrystallized on the spider silk matrix.

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Deer Cave, Gunung Mulu National Park, Sarawak, Borneo (4.056°N, 114.827°E), is known as one of the largest cave passages in the world (Waltham, 2004). The tourist route brings visitors through the large, lower-level, river passage, but the high-level passages in Deer Cave are rarely visited, and are considerably drier than the main passage. The whole cave is well ventilated, with winds changing direction with the time of day. Three very large colonies of the Wrinkle-lipped bat, Chaerophon plicatus, together with smaller populations of at least 11 other bat species (Hall, 1996) and a substantial population of the Black-nest Swiftlet (Aerodramus maximus), occupy the cave. Guano deposits are ubiquitous. This report describes the occurrence of a novel type of gypsum deposit formed on the wall of one of the higher-level passages, related to the presence of nearby guano.

Materials and methods
Material was collected under permit during April 2016, from the cave wall at an elevation of about 1.8m above the soil/dry-guano floor in a high-level oxbow passage above the modern stream in Deer Cave (Fig.1). Initially the sample was photographed in-situ, and then again at the macro-scale, and finally using a scanning electron microscope (SEM) at the Carleton University Nano-Imaging Facility in Ottawa. Identification was by X-ray diffraction (XRD) analysis in the University of Ottawa X-Ray Core Facility.

Microclimatic parameters at the study site were recorded using iButton Hygrochron DS1923-F5 loggers (measuring temperature and relative humidity) and a custom-built anemometer–Arduino logger (measuring windspeed and direction).

Figure 1: Location of the mineral site, Deer Cave, Gunung Mulu National Park. Base map modified from Brook and Waltham (1978).
Figure 2B:
Close-up view of the spiders’ webs and the encrusting crystals, which form a mat that peels off the wall as the weight increases.

Figure 2A:
Location and context of the “arachnite” deposit, on the cave wall just above the head of the figure.
Results

Spiders, particularly a small species of probable Ochyroceratidae (Chapman, 1980), are common in this windy, dry, high-level, passage. Their webs shroud most of the rock surfaces (other than surfaces directly in the path of the strongest winds, where on-going, highly active, condensation corrosion yields clean, white, smooth surfaces). The rock surface on which the spiders build their webs is very much softened (most likely by a combination of condensation corrosion and biogenic activity), to a depth of approximately 1 cm. The surface that presents in the field is a complex of discoloured outermost rock crust (Fig. 2A), plus encrusting webs, plus crystals barely visible to the naked eye, a material that we refer to here as “arachnite”. A combination of the deteriorating rock surface and an increasing weight of encrustation means that the whole complex tends to sag in pendulous gobbets and then peel off intermittently in patches (Fig. 2B), contributing to the fine, dusty accumulations at the sides of the dry guano cave floor. XRD analysis of the material yielded gypsum (CaSO$_4$·2H$_2$O) as the main component.

Gypsum belongs to the monoclinic crystal system and can present in a wide variety of morphologies. These include monoclinic tabular and complex tabular crystals or, as a result of twinning, which is common, swallowtail twins and penetration twins (Freidman, 2017). The crystal habit can be prismatic, acicular, bladed, sometimes bundled in fragile acicular crystals, sometimes as aggregates of fibres, or scales, or grains, sometimes lenticular (flattened lens shapes), or in platy aggregates forming rosettes. When viewed under SEM the “arachnite”, displays many of these different crystal forms and habits.

SEM images reveal two main arrangements. The first comprises delicate acicular and bladed crystals, clumped around and radiating from linear “branches” resembling tiny Christmas trees (Fig. 3A, B, C). Most of the original webs upon which the crystals nucleated are no longer visible, but some bare patches can still be seen. For example, marked by the arrow in Figure 3A and enlarged in Figure 3B, the curved fibres of the spider silk, each of which may have a diameter of only 73–400 nm [1 nm = 1×10$^{-9}$ m] (Anon, 2017) can be seen to be clumped into a ~ 60 micron-diameter bundle like an electrical cable, emergent from its burden of crystals.

The other main arrangement is of lenticular crystals (Fig. 4A, B). These are similar in form to those in desert rose forms of gypsum, the petals made from crystals flattened on the c axis. These are also arranged radiating outwards from a central linear web, but they are more solid in appearance than the Christmas-tree form.

Other crystal habits that can be seen include monoclinic tabular crystals, swallowtail twinned crystals, and prismatic crystals (Fig. 5A, B). The maximum size achieved by any of the crystals is only about 200 μm, and many of the crystals are themselves encrusted with even tinier, second-generation crystals (Fig. 6).

Discussion

The crystals of “arachnite” can only be formed by the evaporation of sulphate-bearing fluids. Temperature and airflow logs at a nearby site show diurnal temperature variations of ~2.5°C amplitude (~4 pm peak to ~7 am trough), and variable but periodically high air flow (median ~ 1.4 m/sec; max 2.8 m/sec). The Melinau Limestone bedrock is highly pure, comprising CaCO$_3$ with less than 1% insoluble residue (Webb, 1982). It is therefore unlikely to be the source of the sulphate component of the gypsum. Rather, the apparent source of the sulphate is the gypsum efflorescence on nearby decomposing guano that is transported as fine dust by the strong air currents.

Figure 3: SEM images of the “arachnite”: “Christmas Tree” form.
A (top): Crystals arranged in whorls around the original spider’s web template. Some of these crystals show a more acicular habit (marked with “A”), some more bladed (marked with “B”). A fragment of the original web that acted as the nucleus for crystal encrustation can be seen as the bundle of fibres marked with an arrow.
B (middle): Enlargement showing details of the web fragment.
C (bottom): Detail of morphology and interconnections of the bladed, twinned crystals.
Gypsum is a common mineral in decomposed guano sequences (Onac and Forti, 2011; Wurster et al., 2015), developing by direct interaction of sulphuric acid with limestone bedrock, the former deriving from the actions of sulphur oxidizing bacteria (Waksman and Joffe, 1922; Okabe et al., 2007). In this case, the fact that the “arachnite” crystals are aligned with the spider silk matrix requires that the crystals form in-situ on the webs, from solution. The mechanism proposed is that condensation water droplets (initially highly undersaturated with respect to gypsum) collect on the webs, trap and dissolve wind-blown gypsum dust, and then re-precipitate it as “arachnite” as changing humidity switches the regime from condensing to mildly evaporative conditions.

Our observations of temperature and airflow were made over 5 days of relatively dry weather; during periods of stormy weather, greater excursions would be expected. With more extreme diurnal shifts in relative humidity, we envisage that the dissolution/re-precipitation process may occur in a single day (condensation from moist, warm air encountering the night-cooled cave walls, followed by mid-afternoon evaporation). The “arachnite” crystals are very small, reaching a maximum size of ~200 microns.

Experiments on gypsum crystallization by Peng et al. (2015) resulted in gypsum crystals up to 300 µm in size forming in just 24 minutes at 25°C, with slightly larger crystals at higher temperatures in similar timeframes (e.g. up to 400 µm at 40°C in 32 minutes). Qian et al. (2012) studied gypsum crystallization from a supersaturated aerosol (using high-speed cameras to record crystal growth) in relation to relative humidity: During the 120 seconds of the experiment, stable gypsum was formed at 81–82% RH, and the crystal growth rate was 0.7–0.8 µm/s. (e.g., rod-shaped crystals of gypsum reached lengths of 17 µm at 105 seconds). These laboratory observations suggest that our proposed mechanism of microcrystal formation on diurnal timescales, in response to varying relative humidities, is entirely feasible.

Figure 4: SEM images of the “arachnite”: “Desert Rose” form. A (top): Platy or lenticular crystals arranged around a linear form (with some of the “Christmas Tree” form visible at top left). B (bottom): Detail of morphology and interconnections of the lenticular crystals. These do not show twinning.

Figure 5: Some of the other crystal morphologies that are shown in the “arachnite”. A (top): Monoclinic Tabular, shown as “MT”, Swallowtail Twin shown as “ST”, plus a little of the spider silk fibres, shown as “SS”. B (bottom): Prismatic habit, marked with arrows.
Conclusion
The formation of distinctive gypsum growths on a spider silk matrix, reported here, adds to the extraordinary variety of known gypsum speleothem forms. This great diversity of form has been attributed to the fact that sulphate ions are rather common in cave contexts, including those in limestone, gypsum and igneous bedrocks, and also to the high reactivity of the sulphate ion (Hill and Forti, 1997). Deposits reported here represent a hitherto unrecognized growth form of gypsum, distinctive for its unique substrate (spider silk), and what we propose to be potentially very rapid formation resulting from diurnal fluctuations in air transport, relative humidity, condensation, dissolution, and re-precipitation.

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References

Figure 6: Lenticular crystals with overgrowths of smaller, second-generation crystals.