A geochemical model for the preservation of insects in the Crato Formation (Lower Cretaceous) of Brazil

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Key words: Crato Formation, Lower Cretaceous, Aptian, Brazil, Insect, Taphonomy.

Abstract

The Crato Formation Lagerstätte of NE Brazil is well known for the exceptional preservation of its fossil assemblage. A wide variety of plants and animals are preserved with a high degree of fidelity, revealing structures of labile tissues at the micron scale, or even smaller. Chemical analyses and descriptions of replacement fabrics observed in insect fossils from the formation are here re-evaluated. Two mineral phases with three distinct fabrics are identified replacing soft tissues: 1) nano-crystalline high-fidelity pyrite replacements of insect epicuticle (pseudomorphed in goethite); 2) pyrite framboids and pseudoframboids obliterating original insect morphology (pseudomorphed in goethite); 3) globular replacements and encrustations of internal labile tissues in calcium phosphate. A revised taphonomic and geochemical model is presented that accounts for both the high fidelity of preservation and the micro-fabrics observed. In addition, we consider aspects of the depositional environment, including substrate consistency, water depth and light penetration. We

conclude that the water depth was likely in excess of 50 m, but within the photic zone, and the sediment was bound by stratified microbial communities.

1. Introduction

The Lower Cretaceous (Aptian) Crato Formation of north-eastern Brazil is a Konservat-Lagerstätte formed in a lacustrine environment (Martill et al., 2007a; Osés et al, 2016; Varejão et al., 2019). It yields a remarkably well-preserved and diverse assemblage of terrestrial, freshwater and possibly anadromous organisms (Davis and Martill, 1999; Martill et al., 2007a). This assemblage includes the oldest gilled mushroom (Heds et al., 2017) as well as arachnids, crustaceans, insects, myriapods, bony fish, anurans, squamates, testudines, crocodilians, pterosaurs and birds, and most major tracheophyte groups known from the Lower to middle Cretaceous (Mohr et al., 2007). Of these, the terrestrial arthropods (insects, arachnids, and myriapods) are particularly well-preserved and important (Barling et al., 2015). Their importance is primarily derived from the mid-Cretaceous (Aptian) age of the formation, which coincides with a major angiosperm radiation, providing a unique window into the evolution of pollination relationships between flowering plants and insects (Hochuli et al., 2006). Consequently, insects of the Crato Formation have been a topic of scientific study for nearly four decades (see Bechly, 2007). Their occurrence was first formally highlighted by Brito (1984) and many insect species were described thereafter (Martins-Neto, 1987; 1988; 1989a; 1989b). A more comprehensive review of the insect fauna, examining approximately 3000 specimens was provided by Grimaldi and Maisey (1990), who noted high relief and the retention of micron-scale cuticular surface details in the fossils (Grimaldi and Maisey, 1990, fig. 5). Their findings were corroborated by Bechly (1998), who also observed micron-scale preservation of cuticle replaced by goethite, along with the presence of internal tissues, stomach and gut contents, high relief, and structural colour. Despite three-and-a-half decades of study, the mechanisms of this exceptional preservation remains poorly understood.

1.1. Previous taphonomic investigations

Several authors have investigated the mechanisms that control the exceptional preservation of the Crato Formation insects. Firstly, Grimaldi and Maisey (1990) identified goethite as the preserving mineral (with no original organic materials preserved) and hypothesised that a well-oxygenated freshwater palaeoenvironment was required for this mode of preservation.

Further discussion by Menon and Martill (2007) noted that two preserving mineral phases are present; an orange/brown hydrated iron mineral (goethite) and a black “carbonaceous” material associated with pyrite. These mineral phases were correlated with overburden thickness and degrees of weathering (Menon and Martill, 2007). They concluded that the goethite phase represents a deep, slow, and in situ weathering product of the black “carbonaceous” mineral (pyrite), resulting in goethite pseudomorphs of original pyrite fabrics. Other, rarer preserving minerals were also reported: phosphatised muscle tissues, voids infilled with clear calcite, and rarer silicified concretions around fossils (Menon and Martill, 2007). These authors inferred that sulphate reducing bacterial mats played an important role in this preservation, dominating the microbial community at the anoxic, sulphur-rich, and hypersaline sediment-water interface. Menon and Martill (2007) also highlighted the unusual taxonomic composition of the insect fauna, noting the abundance of strong flying (typically in winds exceeding 5 m/s (Corbet, 1999)), burrowing, and cryptic taxa. Additionally, there appears to be little selection for the robustness of the insect assemblage, nor any evidence for mass mortality events (Menon and Martill 2007) There is, however, an obvious bias against small (< 5 mm) insects, and they are extremely rare compared to larger ones. The preservational fidelity of the smaller insects is the same as the larger insects and, consequently, large, small, delicate, and robust taxa are all preserved with high-fidelity, possibly as a consistent influx of carcasses during seasons of heavy rain (Menon and Martill 2007).

More recently, several studies have investigated the preservational processes preserving arthropods in the Crato Formation . One goethite preserved orthopteran specimen analysed by Delgado et al.

(2014) indicated the presence of Fe and O, as well as lesser amounts of P and Mg. They identified pseudomorphed pyrite framboids as the replacement fabric, differing in size between external cuticle and internal tissues, with internal ones being considerably smaller (Delgado et al., 2014, fig. 6). They also reported the presence of mineralised extracellular polymeric substances (EPS).

Subsequently, Barling et al. (2015) highlighted the highest-fidelity preservation of Crato Formation insects, including retention of submicron-scale cuticular surface details and the preservation of labile internal organs. These authors provided further examples of pseudomorphed pyrite framboids and pseudoframboids of varying size and surface organisation (Barling et al., 2015, fig. 14), noting that some of the internal tissues were preserved in calcium phosphate. The external replacement textures observed by Barling et al. (2015) differed from those of Delgado et al. (2014). Barling et al. (2015) also noted submicron-scale, near-perfect replications of the epicuticle, with fragile structures retained (Barling et al., 2015, fig. 8), whereas these were obliterated by pseudomorphed framboids in the specimen examined by Delgado et al. (2014, fig. 6).

A comprehensive analysis of Crato Formation insect preservation was undertaken by Osés et al. (2016). They briefly summarised previous taphonomic studies, undertook further analyses, and presented a model for the preservational process. Several fossils were subject to energy dispersive X-ray analysis, energy dispersive X-ray fluorescence, soft X-ray spectroscopy, particle induced X-ray emission, and Raman spectroscopy (Osés et al., 2016). They concluded that the decay of labile tissues, combined with the metabolic activities of sulphate reducing bacteria, resulted in hydrogen sulphide saturation and framboidal pyrite precipitation. Framboid size was limited in some areas by the rapid exhaustion of labile tissues and limited ion availability, although some ions diffused into the carcass through cuticular micro-cracks (Osés et al., 2016). This resulted in finer-grained framboid formation within the carcasses, compared to the coarser framboids of the cuticle, corroborating the observations of Delgado et al. (2014). They further suggested that carcass compaction was prevented by early pyritization and the inherent structural integrity of the insects, and they

confirmed that the highest fidelity internal labile tissues were preserved in early diagenetic calcium phosphate (Osés et al., 2016). In the model presented by Osés et al. (2016), sulphate ions originated from 'evaporites', however it is unclear which sediments they are referring to and there appears to only be evidence for short-lived halite growth within the Crato Formation (Martill et al., 2007b).

Most recently, the preservational mechanics of Crato Formation arthropods was reassessed with a focus on the role of microbial mats (Varejão et al., 2019). This investigation examined two decapod shrimp fossils as examples of arthropod preservation and proposed several key differences from previous studies. Varejão et al. (2019) noted that there are three distinct sedimentary intervals and that fossils of some organisms are only found at certain levels of those intervals. For example, while fish and plant fossils occur throughout the Nova Olinda Member, insects are restricted to the top of the uppermost interval whereas terrestrial vertebrates are only found in the base of the lowermost interval. These intervals are defined by the presence of different sizes of stromatolites and fossils were assigned based on microfacies analysis (Varejão et al., 2019). The abundance of stromatolites is also reaffirmed (see Warren et al., 2017) and their impact on the preservation of arthropods is discussed (Varejão et al., 2019). Varejão et al. (2019) suggest two key differences from previous models: that the Crato palaeolake was shallower than previous descriptions and that its bottom waters were oxygenated. These are supported by the presence of stromatolite structures. The justification for allowing an oxygenated bottom water environment, while allowing for pyrite formation (an anoxic process, discussed below in section 5.3.) is that carcasses were entombed by the microbial mats and transitioned through several geochemically distinct microbial zones (Varejão et al., 2019).

1.2. Justification for a new preservational model

Hypotheses and a model for the preservational mechanisms of the Crato Formation entomofauna have previously been presented. While the model by Osés et al., (2016) adequately describes the

sedimentological, bottom water, and microbiological conditions required for this mode of preservation, it does not explain all of the preservational fabrics observed in the insect fossils (Barling et al., 2015). Osés et al. (2016) reported coarser framboidal pyrite replacing the exterior-most cuticle and suggest that areas of high-fidelity cuticle replacement are a consequence of the 'close packing' of these framboids. Such an interpretation is also supported by Delgado et al. (2014). Conversely, Barling et al. (2015), and data herein suggest that three distinct fabrics of preservation are discernible in most specimens and consequently a revised preservational model to explain these differences is required. Additionally, the model presented here expands further on the suggestion by Varejão et al. (2019) that carcasses were entombed by microbial mats and underwent several stages of mineralisation, but offers a much more detailed description of the mineralisation processes and which tissues they affect.

2. Locality and geological background

The Crato Formation crops out along the flanks of the Chapada do Araripe, a ~150 km plateau located along the borders of the states of Ceará, Pernambuco, and Piauí (Fig. 1). It is especially well exposed in stone quarries between the towns of Nova Olinda, Santana do Cariri, and the farmstead of Tatajuba. The fossils described here all come from the numerous stone quarries in this region (see Martill and Wilby (1993) for details).

The formation consists of four members; the Nova Olinda Member, the Caldas Member, the Jamacaru Member, and the Casa de Pedra Member (Fig. 2; Martill et al., 2007a). Of these, the lowermost Nova Olinda Member is the Konservat-Lagerstätte that has been studied extensively (Martill et al., 2007a and the references therein). There are alternative stratigraphic schemes, but we adopt the system proposed by Martill and Heimhofer (2007) and expanded upon by Heimhofer et al. (2010). It rests disconformably on the Barbalha Formation in the east and on red beds of an

unnamed formation in the north west of the outcrop. In a few places, it onlaps with topographic unconformity on the Cariri Formation (age uncertain) and Precambrian metamorphic basement.

The Nova Olinda Member is up to 13 m thick with two distinguishable macrofacies: finely laminated rusty-brown clay-carbonate rhythmites, with varying amounts of organic matter, and a laminated limestone macrofacies that resembles a typical 'plattenkalk' (Neumann et al., 2003). These can be further separated into four distinct microfacies, with the clay-carbonate rhythmites separated into rhythmic and nodular microfacies, and the laminated limestone separated into planar laminated and crustiform microfacies (Catto et al., 2016). All samples analysed here are from the laminated limestone macrofacies, and planar laminated microfacies, the latter characterised by discrete interlocking calcite rhombohedra (Fig. 3: A and B). In areas of increased overburden, the limestones appear blue-grey (unweathered) and yield black fossil arthropods, whereas in areas of less overburden, the limestones are pale-cream (weathered) and yield orange/brown goethite fossils, a consequence of differing degrees of weathering (Menon and Martill, 2007). It has been suggested recently that the different colours reflect different sedimentary regimes (Osés et al. 2017; Oliveira, 2018), with the blue-grey coloured strata deposited in the methanogenic zone, while the pale cream coloured strata was deposited in the sulphate reduction zone. This hypothesis is untenable as strata can be traced laterally from cream coloured to blue-grey coloured in quarries over very short distances. The colour change often reflects the topographic surface and is clearly a consequence of modern weathering.

3. Material and methods

3.1. Materials

A total of 135 fossil insect specimens were analysed, kindly donated by J. Wohlrabe. The collection has a relatively low taxonomic diversity and is biased towards members of Blattodea and Orthoptera (Fig. 4), when compared with previous studies (see Menon and Martill 2007). All specimens, except one are stored at the University of Portsmouth (Portsmouth, UK). One specimen (SMNS 70092) has been deposited at the Staatliches Museum für Naturkunde Stuttgart (Stuttgart, Germany) (see Barling et al., 2013).

Most specimens are on angular slabs of laminated limestone, approximately 10 – 15 cm in diameter. Excess rock was cut away with a diamond circular rock saw, lubricated with deionised water, leaving approximately 0.5 – 1 cm of limestone matrix remaining around the fossil. Trimming enabled specimens to fit into analytical equipment (e.g. the scanning electron microscope chamber), and also allowing them to be stored closely packed in PELCO® SEM stub containers.

3.2. Methods

Most of these specimens were prepared for scanning electron microscopy and energy dispersive X-ray spectroscopy via a refined version of the resin transfer technique of Barling et al., (2019). Two specimens were selected for destructive X-ray diffraction spectroscopic analysis.

3.2.1. Scanning electron microscopy

Over 3,500 scanning electron microscope (SEM) images were captured, depicting the micron-scale preservational fabrics. Three SEM units were used during this project: a JEOL JSM-6100 Scanning Microscope, a JEOL JSM-6060LV Scanning Electron Microscope and an EVOMA10 Zeiss Scanning Electron Microscope with a variable pressure environmental chamber. Working distances varied between 11 mm and 32 mm, with most specimens examined at approximately 13 mm working

distance. This was varied based on the fragility of each specimen and its topographic relief. The majority of specimens were examined at 16 kV, although a broad range of voltages (8 kV – 20 kV) were used with the selected voltage depending on the fragility of the specimen and how effectively it dissipated charge. Only the most durable specimens were examined at 20 kV while only the most fragile below 10 kV. For most specimens, the spot size (probe diameter) was kept at the default 60 nm. Probe currents varied between 5 pA and 200 pA, allowing for images of uncoated specimens to be captured at magnifications up to 85,000 ×.

3.2.2. Energy dispersive X-ray analysis

Energy dispersive X-ray analysis (EDX) was used to determine the elemental composition of the fossils and surrounding matrix. Six specimens were analysed with EDX, but the spectra of only one (specimen UOP-PAL-NB104) is presented here as it possesses every mineral phase and fabric observed and was meticulously prepared (by Florence Gallien) to provide clear examples of each.

To achieve the best results for EDX analysis, specimens were polished to a flat surface (CFAMM, 2018). However, specimens were prepared using the resin transfer technique (Barling et al., 2019), and left with a fragile surface with high topographic relief. Consequently, there is some inherent error due to topographical variations of the samples, but this was necessary to distinguish replacement fabrics and the EDX results still provide useful and interpretable information.

Acceleration voltage should, ideally, be no less than half of the highest excitation energy of any elements present (CFAMM, 2018). Fe was the highest excitation energy element present (7.11 keV), and so the acceleration voltage was no lower than 14.22 keV where possible (CFAMM, 2018). An acceleration voltage of 20 keV was used for analysing specimen UOP-PAL-NB104.

Several specimens (including UOP-PAL-NB104) were gold sputter coated to enable sharper SEM imaging. The energy dispersive X-ray spectra of gold overlap with spectra for rubidium and sulphur,

making it difficult to discern the abundance of those elements in the specimens (Newbury, 2009), and so caution should be used when identifying them. It should be noted that rubidium is not expected in Crato Formation fossils, whereas sulphur should be present due to the presence of pyrite (Grimaldi and Maisey, 1990; Menon and Martill, 2007).

3.2.3. X-ray diffraction spectroscopy

Two samples were subject to X-ray diffraction (XRD) in a PANalytical X'PERT-3 diffractometer. Samples of cuticle were taken from each specimen, ground to a powder, homogenized, randomly orientated on a glass slide, packed into a sample container, and irradiated with X-rays (Klug and Alexander, 1974). This resulted in constructive interference, which, when Bragg's Law was met, could be plotted on a X-T intensity plot diffractogram (Klug and Alexander, 1974). Each diffractogram was compared against a database of standard reference patterns, allowing for mineral species identifications. Any mineralogies that comprise less than two percent of the sample went undetected (Klug and Alexander, 1974).

4. Results

4.1. Preservation fabrics

Crato Formation insects are exceptionally well-preserved but, when scrutinised closely, three discernible fabrics of preservation are identifiable, each with distinct preservational fidelities.

The dominant fabric of preservation (of rusty-coloured specimens) is goethite pseudomorphs of framboidal and pseudoframboidal pyrite (Fabric 1) (Fig. 3: C and D; Barling et al., 2015; Osés et al., 2016). This replaces much of the internal tissue, most of the exo- and endocuticle, and some areas of

epicuticle (Fig. 3: C and D). These replacement structures are remarkably variable in size and surface organisation, but are typically 10 μm in diameter (range = < 1 μm to 20 μm) (Fig. 3: C and D; Barling et al., 2015). They are sometimes hemi-spherical (having grown against a surface), appear to be hollow, and vary in surface organisation (Fig. 3; Barling et al., 2015). In some parts of the fossil, they are fused or inter-grown to form a single granular-to-globular layer (y in Fig. 3: E – G), while in other areas of the same fossil they are much larger and composed of coarser granules (y in Fig. 3: H). For a full description and further examples of the variability of this fabric, see Barling et al. (2015). This fabric is not seen in the ‘black’ Crato Formation insect fossils from the ‘blue’ limestone. Instead, those fossils possess a smooth-to-globular surface that appears to retain little-or-no microscopic detail or discernible crystallography (Fig. 5). Aside from the gross shape and volume of the insect, there does not appear to be any cuticular morphology preserved. This surface is preserved in pyrite and/or gypsum (see Section 4.3. for further information) and may represent a ‘blanket’ of amorphous organic material (AOM) covering the cuticle.

In rust-coloured goethitic specimens, a second distinct fabric is present (Fig. 3: E – H; Barling et al., 2015). Where the external-most cuticular features (epicuticle) are preserved with remarkable fidelity, original anatomical morphology is replicated at the sub-micron scale by nanometre-size crystallites (Fabric 2) (Barling et al., 2015; x in Fig. 3: E – H). This preservation is almost exclusive to the epicuticle and other external features, including setae and ommatidia, but is found rarely extending into the exocuticle (Barling et al., 2015, fig. 13: F). Where present, there is a clear distinction in fidelity of preservation and grain-size from that of the pseudomorphed framboidal fabric (Fig. 3: C – H).

While many of the contacts between fabrics 1 and 2 are sharp, there are areas where Fabric 2 appears to transition gradually into a globular texture, then into Fabric 1 (e.g. Barling et al., 2015, fig. 14: A). These fabric transitions are associated with cuticular perforations (e.g. setae and spiracles) or cuticular damage (Barling et al., 2015, fig. 14: A; Fig. 6: A and B).

An additional, though less commonly occurring fabric is that of calcium phosphate impregnations and globular encrustations (Fabric 3) (Fig. 6: C – F). This fabric exclusively replicates internal labile ‘soft’ tissues. It does so with moderately high-fidelity, preserving individual muscles and muscle fibres (Fig. 6: E and F), but largely obscures their surface morphology with globular encrusting mineralisations (Fig. 6: F). These fabrics were also highlighted by Barling et al. (2015) and Osés et al. (2016). Calcite infills and cements are also common in these fossils (Fig. 6: D(x)).

4.2. Elemental analyses

Energy dispersive X-ray analyses (EDX) presented here corroborate previous studies (Grimaldi and Maisey, 1990; Bechly, 1998; Menon and Martill, 2007; Barling et al., 2015; Osés et al., 2016). Here, iron and oxygen are identified as the dominant elements preserving insect tissues (Fig. 7), and their occurrence corresponds with fabrics 1 and 2 (Figs. 7 and 8). Calcium and phosphorous are the next most abundant elements observed replacing tissues (Fig. 7). These are restricted to Fabric 3 and confirm previous suggestions of calcium phosphate preservation (Fig. 8; Barling et al., 2015; Osés et al., 2016). EDX spectra also highlight the presence of calcite void infills (Fig. 8: spectrum 1).

4.3. Mineralogy

Two specimens (UOP-PAL-NB114 and UOP-PAL-NB111) were selected for destructive mineralogical analysis (X-ray diffraction). Specimen UOP-PAL-NB114 is preserved in the typical rusty orange/brown colour, whereas specimen UOP-PAL-NB111 is from the ‘blue’ limestone and is preserved with a black colouration. A portion of cuticle and internal tissues were taken from each specimen and ground to a powder.

X-ray diffraction of specimen UOP-PAL-NB114 revealed goethite as the dominant replacing mineral, with calcium carbonate as calcite also identified (likely due to contaminant fragments of the

surrounding matrix being incorporated) (Fig. 9). Specimen UOP-PAL-NB114 was selected for destructive analysis on the basis of its abundance of fossil tissues, but otherwise relatively poor preservation. The analysis did not detect calcium phosphate minerals and it is likely that this specimen did not possess internal soft tissue preservation. However, based on previous investigations (Osés et al., 2016), the calcium phosphate mineral is suggested to be apatite. X-ray diffraction of specimen UOP-PAL-NB111 revealed abundant calcite (again, mostly likely due to matrix contamination), but identified pyrite and an uncertain identification of gypsum as the dominant preserving minerals (Fig. 9).

5. Discussion

5.1. Fabrics and preserving minerals

Three fabrics of preservation were identified: 1) pseudomorphed framboids, 2) high-fidelity nanocrystalline replacement, and 3) globular internal soft-tissue incrustation and replacement. The chemical and mineralogical analyses presented herein further confirm that, in the 'orange/brown' Crato Formation fossil insects, these fabrics are composed of two primary preserving minerals: goethite, which corresponds to fabrics 1 and 2, and calcium phosphate (?apatite), which corresponds to fabric 3.

Fabric 1 is undoubtedly the dominant fabric, replacing the vast majority of insect tissues. Where it does so, fidelity is poor, mostly obliterating the original morphology (Fig. 3: C and D). This is due to the initial replacing crystals being coarser grained (~1 µm diameter) cubic pyrite, growing as framboids and pseudoframboids. These were later pseudomorphed by goethite, giving rise to the globular subspherical fabric observable today.

313 Despite Fabric 2 sharing the same mineral phase transitions as Fabric 1 (weathering from pyrite to
314 goethite), it differs considerably in preservational fidelity. Although this fabric is also a result of
315 pyrite pseudomorphing, initial pyrite crystal growth was non-framboidal and probably
316 nanocrystalline (approximately between 10 – 100 nm). Nanocrystals impregnated the epicuticle,
317 replicating the original morphology at the submicron scale (x in Fig. 6: E – G). This high-fidelity
318 preservation is non-ubiquitous and in some fossils the epicuticle is replaced by coarser grained pyrite
319 that traces topographic outlines of the underlying pseudomorphed framboids. This was observed
320 and described by Delgado et al. (2014) and Osés et al. (2016), and is the basis for their interpretation
321 of these fabrics being the same.

322 The globular encrustations and replacements by calcium phosphate of Fabric 3 strongly resembles
323 ‘Orsten’-type calcium phosphate replacement (Maas et al., 2006; Eriksson et al., 2012; Wilby pers.
324 commu., 2016). Previous investigations have suggested that abundant bony fish skeletons in the
325 surrounding sediment are necessary for calcium phosphate replacement, but it is now known that
326 arthropod carcasses can supply enough phosphate for mineralisation of their own tissues (Briggs et
327 al., 1993; Briggs and Kear, 1994; Martínez-Delclòs et al., 2004). Nevertheless, this mode of
328 preservation is bacterially mediated and requires ‘active sites’ for precipitation (Briggs et al., 1993;
329 Wilby and Briggs, 1997; Martínez-Delclòs et al., 2004; Maas et al., 2006; Osés et al., 2016). These
330 ‘active sites’ are created in tissues by their decay (Martínez-Delclós and Martinell, 1993; Briggs,
331 1995; Kaposos and Koutsoukos, 1999). Calcium phosphate preservation is also known to be
332 widespread within the Crato Formation, preserving vertebrate soft tissues (Davis and Martill, 1999).

333 In areas where Fabric 2 transitions to a globular texture, and then to Fabric 1 (Figure 6: A and B), this
334 likely represents preservational fidelity loss caused by ongoing decay during fossilisation. This decay
335 would have been facilitated by nutrients escaping from decaying internal tissues, which explains the
336 textures’ association with cuticular perforations (like setae bases and spiracles). Alternatively, these

same seeping nutrients instead changed the localised microgeochemical conditions around the perforations and this alteration in fabric is a result of incomplete mineralisation rather than decay.

5.2. Differences from previous models

The proposal of three distinct fabrics of preservation differs from the model presented by Osés et al. (2016). In their model, the high-fidelity replication of the epicuticle (Fabric 2 here) is proposed to be a result of the 'close packing' of pseudomorphed framboids (Fabric 1 here). Here, the justification for considering these to be separate fabrics is as follows:

1) The size of the crystallites differs between the two fabrics, with the high-fidelity fabric (Fabric 2) composed of nano-crystallites and the pseudomorphed framboids and pseudoframboids (Fabric 1) composed of much larger submicron-crystallites.

2) The high-fidelity fabric (Fabric 2) only replaces the epicuticle (and very rarely exocuticle), whereas the pseudomorphed framboid and pseudoframboid fabric (Fabric 1) replaces internal tissues and subsurface cuticular layers (exo- and endocuticle).

3) Where these fabrics merge or intermingle, they do so in areas where the epicuticle has been 'degraded' through decay or incomplete mineralisation.

With these fabrics clarified, a refined model is presented to account for the chemical conditions required to produce this additional fabric. However, the presence of framboidal (and pseudoframboidal) pyrite pseudomorphs, and 'Orsten'-type preservation means that any proposed taphonomic model must explicitly include the conditions necessary for the formation of these fabrics. Consequently, the processes of pyrite framboid formation and 'Orsten'-type preservation must first be outlined.

The model presented here also differs from the preservational model presented by Varejão et al. (2019). Their model does not address the microfabrics of preservation of the arthropod tissues, but

instead examines the microfabrics adjacent to the carcasses generated by the entombing microbial mat.

5.3. Pyrite framboid formation and definition

Many studies on fossil soft tissue preservation refer to globular, grape-like pyrite grains as ‘framboids’ (Sawlowicz, 2000). However, true pyrite framboids have the distinct definition of ‘microscopic spheroidal to sub-spheroidal clusters of equidimensional and equimorphic microcrystals’ (Ohfuji and Rickard, 2005). They may have disorganised, partially organised, or regularly organised interiors and exteriors, and are typically ~10 µm in diameter, but infrequently can range between 1 µm – 250 µm (Kribek, 1975; Ohfuji and Rickard, 2005, fig. 1). Pseudoframboids are simply framboids that lack the internal microcrystal structure of ‘true’ framboids (Ohfuji and Rickard, 2005). Instead, they are filled by a single larger massive pyrite crystal or are hollow. As many of the pseudomorphed framboid structures observed in these fossils are hollow, they would have been classified as pseudoframboids, prior to their pseudomorphing in goethite.

Pyrite formation has been, and continues to be, an important topic of geochemical investigation (Ohfuji and Rickard, 2005; Wu et al., 2012; Vietti et al., 2015 and references therein). Low temperature (< 100°C) pyrite geochemistry is complex, polyphase, and extremely oxygen-sensitive (Wolthers et al., 2003; Ohfuji and Rickard, 2005). Pyrite is typically stable at surface temperatures and pressures but can be metastable depending on the size and nature of its crystals (Berner, 1970; Ohfuji and Rickard, 2005; Rickard, 2012). A series of metastable non-pyritic iron sulphides must precipitate before pyrite can form (Berner, 1970; Vaughn and Craig, 1978). These include non-crystalline ‘amorphous iron monosulphide’ (FeS, sometimes called disordered mackinawite), mackinawite ((Fe,Ni)S_{0.9}), and greigite (Fe₃S₄) (Skinner et al., 1964; Jeong et al., 2008). Of these, amorphous iron monosulphide is the first phase to precipitate during the reaction between Fe²⁺_{aq} and S²⁻ under ambient conditions, and does so during early diagenesis just below the sediment

surface (Berner, 1970; Lennie and Vaughan, 1996; Wolthers et al., 2003). Two different reactions can occur depending on the acidity and temperature of the pore water:

As these reactions progress in a closed system, the pH of the host solution decreases, eventually causing them to stop (Wolthers et al., 2003; Ohfuji and Rickard, 2005). Consequently, time is also a controlling factor. These reactions form 'subparticles' (nano-crystals) between 20 – 400 nm in diameter, which are difficult to discern, even under high magnification SEM (Wolthers et al., 2003). After initial precipitation of an iron monosulphide phase, it reacts with elemental sulphur (S_0), polysulphides (H_2S_n), or hydrogen sulphide (H_2S) to form pyrite (Wu et al., 2012). If elemental sulphur and S^{2-} ions are present, both will react (Ohfuji and Rickard, 2005). Rickard (1997) simplified the typical pyrite formation reaction to:

More recent studies have suggested that an intermediate phase of greigite may form during the conversion of mackinawite (disordered or ordered) into pyrite (Hunger and Benning, 2007). Several complex steps occur that can be summarised in the following reaction:

In the equation above, mackinawite can alternatively transform into pyrite via iron loss (as oppose to sulphur gain) (Butler and Rickard, 2000; Hunger and Benning, 2007). Iron loss in this manner results in significant volume shrinkage (up to 41.7% iron loss), which would drastically alter fossil tissues if it occurred in them.

Most experiments indicate that pyrite supersaturation is necessary for framboids to form, along with anoxia and abundant sulphate reducing bacteria. However, Vietti et al. (2015) replicated framboid-like structures (interpreted as precursor 'protoframboids') in oxic experimental conditions, indicating that the controls for framboid formation occur on the microgeochemical scale, rather than at the gross sedimentary scale. This explains why two different pyrite replacement fabrics can occur within a single fossil, as observed in the Crato Formation.

5.4. 'Orsten'-type preservation

The Cambrian Orsten Konservat Lagerstätte of Öland, Sweden preserves arthropod fossils exceptionally (Müller and Walossek, 1986). In a preservational mode dubbed 'Orsten'-type preservation, labile soft tissues are replicated in high-fidelity via encrustation and replacement by calcium phosphate with a sub-micron scale granular coating (Maas et al., 2006, fig. 1; Eriksson et al., 2012). This type of preservation is attributed to bacterially-mediated calcium phosphate precipitation (Briggs et al., 1993; Martínez-Delclòs et al., 2004; Maas et al., 2006). Arthropod carcasses can supply sufficient phosphate for mineralisation of their own tissues during decay (Briggs and Kear, 1994), however some authors propose that a build-up of phosphates in the sediment, rather than in the carcass, is also required (Briggs et al., 1993; Martínez-Delclòs et al., 2004). Increased sedimentary phosphate levels can be achieved by the dissolution of tissues and skeletons of bony fish carcasses within the sediment (Briggs et al., 1993; Martínez-Delclòs et al., 2004). Although bony fish are highly abundant in the Crato Formation (Martill et al., 2007a), it is unlikely that their skeletons provided a source of phosphorous for the fossilisation of the insect labile tissues as there is no evidence for the dissolution of their bones (Davis and Martill, 1999). Only a small amount of the insect internal tissues are preserved in calcium phosphate and the carcasses were likely chemically 'isolated' from the remaining sediment (discussed further below). As such, the

phosphate replacing these tissues likely came from the decay of adjacent tissues in the same carcass.

5.5. Depositional environment and substrate consistency

The depositional and sedimentary conditions of the Nova Olinda Member of the Crato Formation are a key component of any taphonomic model. Previous authors have suggested that the top of the sediment was 'soupy' (Martill, 1993), allowing carcasses to gently sink into it. This is supported by the presence of ripple and tearing structures, soft sediment deformation, (Martill et al., 2007b; Heimhofer et al., 2010), and the unusual vertical orientation of some arthropod carcasses (Varejão et al., 2019). More recent investigations have alternatively suggested that the extensive nature of the microbial mat and a lack of bottom water currents instead allowed for carcasses to be preserved in vertical orientation (Varejão et al., 2019). In this case, the mat presumably was a 'sticky' substrate, rather than a 'soupy' one.

Another important depositional aspect is the oxygen levels at the sediment-water interface and within the sediment. Previous authors have suggested that the water column was stratified in terms of oxygen levels (Heimhofer et al., 2010; Barling et al., 2015; Osés et al., 2016). However, the identification of benthic microbes as the origin for the limestone (Catto et al., 2016) indicates that photosynthesis was occurring at the sediment-water interface and it has now been suggested that the lower water column was oxygenated (Varejão et al., 2019). Initially, this appears inconsistent with the widespread replacement of fossils by pyrite, which is an extremely oxygen-sensitive process (Ohfuji and Rickard, 2005), but further considerations suggest that the microbial mat itself was stratified, allowing for pyrite formation within an anoxic sulphate-reducing bacterial layer at the bottom of the community (Varejão et al., 2019). Despite the presence of oxygen, the depositional environment was still too hostile for grazing metazoans. This is because the bottom waters were

sufficiently hypersaline to induce halite crystal growth (Martill et al., 2007b). The importance of a stratified microbial mat is supported here.

The confirmation of photosynthesis at the sediment-water interface warrants further consideration, as it highlights the question of water depth. Varejão et al. (2019) suggested that the Crato Formation palaeolake was relatively shallow, however the absence of sedimentary storm structures indicates that deposition occurred at least below storm wave base. Consequently, deposition must have occurred between 50 - 200 m in depth, a depth that is below storm wave base, but still allows for abundant photosynthesis to occur (Genkai-Kato et al, 2003).

5.6. A new preservational model for Crato Formation fossil insects

The observations and analyses presented here have established that a revised preservational model is required for the Crato Formation insects. Below, a refined model is presented primarily as a two-part figure (Figs. 10 and 11) separated into six simplified sections (A-F), but a written narrative is also provided.

Fig. 10: A. The Crato lake/lagoon waters were supplied with abundant dissolved ions by a restricted connection to the south Atlantic (Ponte, 1992; Martill et al., 2007a; Neto et al., 2013). Tributaries and small-scale flash floods also provided soil-derived ions from the hinterland (Martínez-Delclòs et al., 2004; Pearlman, 2016). The restricted oceanic connection limited water exchange and the hot arid climate concentrated ions via evaporation, resulting in hypersaline bottom waters (Martill et al., 2007b). The insect carcasses arrived at the site of deposition and were entombed rapidly by an extensive photosynthetic microbial mat and partially soupy sediments (Martill, 1993; Martill et al., 2007b; Heimhofer et al., 2010; Varejão et al., 2019). Due to the high salinity, no scavenging or burrowing metazoans were present, and the sediment was dominated by microbial organisms (Siegrist et al., 1999; Varejão et al., 2019). Minor decay that had already begun at the surface in

many carcasses was greatly slowed by the high salinity of the lake/lagoon floor environment (Allison and Briggs, 1993; Holliger et al., 1998; Siegrist et al., 1999).

Fig. 10: B. The entombing photosynthetic microbial mat generated isolated microgeochemical environments that favoured mineralisation. The minor decay of the most labile 'soft' tissues liberated ions and created 'active sites' for mineral precipitation (Briggs et al., 1993; Maas et al., 2006). This allowed for calcium phosphate (apatite) to precipitate, encrusting and replacing some of the remaining internal soft-tissues (Kapolos and Koutsoukos, 1999). This resulted in fossilisation similar to the 'Orsten'-type preservation of Sweden (Fabric 3) (Martill et al., 1992; Allison and Briggs, 1993; Maas et al., 2006; Eriksson et al., 2012; Barling et al., 2015; Fig. 6: C – F). Despite the abundance of bony fish in the surrounding sediment, extensive phosphatisation was prevented due to carcass 'isolation' by the microbial mat. Later, precipitation of calcium carbonate allowed for some voids to be infilled by calcite (also see Fig. 11).

Fig. 10: C. As the mat continued to overgrow the carcass, the dominant microbial activity transitioned from photosynthesis to sulphate reduction (Varejão et al., 2019). The metabolic activities of these reducing bacteria concentrated ions, forming a geochemical gradient within the epicuticle of each carcass which, in turn, stimulated mineral precipitation (Berner, 1981; Sørensen, and Jørgensen, 1987; Allison, 1988; Henrichs, 1992; Aller, 1982; Simon et al., 1994). Specifically, sulphate ions present in the hypersaline bottom waters were reduced by the microbial mat, generating hydrogen sulphide (Briggs and Kear, 1994; Chester and Jickells, 2012). This combined with aqueous Fe^{2+} present within the sediments (from decaying photosynthetic microbes), forming iron monosulphide (Chester and Jickells, 2012). Initial precipitation was amorphous iron monosulphide (disordered mackinawite), which then stabilised to nanocrystalline non-framboidal iron monosulphide/greigite, resulting in impregnation of the epicuticle by nanocrystals (Fabric 2) (Sagemann et al., 1999). In some cases, impregnation of the cuticle continued deeper into the carcass, resulting in slower growing, larger (but still submicron-size), crystals (Aust, 1972). These

replicated the exocuticle in a similar manner to the epicuticle, but with a reduced fidelity and thus preserving minimal internal structure (Barling et al., 2015, fig. 13: F). Iron monosulphides also continued to diffuse into the carcass.

Fig. 11: D. Continued bacterial sulphate reduction formed more hydrogen sulphide, which continued to react with Fe^{2+} , forming more iron monosulphide. By the time the remaining interior of the carcass began to mineralise, iron monosulphides had reached supersaturation, resulting in the formation of precursor structures to pyrite framboids and a different replacement fabric (Fabric 1) (Berner, 1970; Vaughn and Craig, 1978; Vietti et al., 2015). As with the nanocrystalline impregnation described above, these crystallites would have initially precipitated as an amorphous iron monosulphide (disordered mackinawite), which then stabilised (into either ordered mackinawite or greigite, or both) (Berner, 1970; Ohfuji and Rickard, 2005; Hunger and Benning, 2007; Wu et al., 2012). These precursor 'protoframboids' (Vietti et al., 2015) varied in size depending on the unique microgeochemical environments within each insect carcass/tissue (Barling et al., 2015, fig. 14). In some cases, they grew against the internal surface of the already mineralised epicuticle, forming hemispherical-protoframboids (Barling et al., 2015, fig. 12: F). The formation of these protoframboids obliterated any remaining internal morphology that had not already been replicated in calcium phosphate or nanocrystalline iron sulphides. Ions diffused into the carcasses through cuticular spiracles, damaged setal bases, and cuticular perforations caused by decay.

Fig. 11: E. As bacterial sulphate reduction continued, the pH within the carcass decreased (Briggs and Kear, 1994) and the aqueous Fe^{2+} was eventually depleted. This resulted in a transition from reactions that form amorphous iron monosulphide to those that form metastable crystalline iron sulphides (ordered mackinawite and pyrite) (Vaughn and Craig, 1978; Wolthers et al., 2003; Ohfuji and Rickard, 2005). The epicuticle (and in some places exocuticle) that had already been mineralised by nanogranular iron monosulphides was replaced by nanocrystalline pyrite (or possibly metastable mackinawite in some places), with a minimal loss of fidelity. Within the carcass, pyrite overprinted

its precursor phases in coarser crystals. Conditions were not appropriate for the generation of true
framboids, resulting instead in pseudoframboids, identified by their hollow interiors (Fig. 3: H)
(Barling et al., 2015, figs. 12: E and 14: C).

Fig. 11: F. Many millions of years later (approximately 112 million years (Coimbra et al. 2002)),
overburden was removed from the sediment, exposing it to oxygenated meteoric water. This
allowed for slow in-situ weathering, replacing the pyrite and any other iron sulphide phases with
goethite (Menon and Martill, 2007). The pyrite replacing the epicuticle (and in some places
exocuticle) was pseudomorphed by nano-crystalline goethite, preventing a loss of fidelity. The
internal pseudoframboids were also pseudomorphed in goethite. However, their coarser cubic
crystal structure was replaced by aggregates of micro- and nano-crystals, giving rise to the
preservational fabric observed today.

5.7. Model limitations

The model outlined above explains the diagenetic pathways that preserve the Crato Formation
insects as high fidelity fossils. Subtle differences of preservation style, e.g. size of pseudoframboids,
degree of mineral replacement, and mineralising textures probably reflect subtle variations in micro-
geochemical environments where Eh, pH, ion concentration and substrate composition of the insect
tissue vary within (probably) very narrow parameters.

Each insect carcass likely contained an array of micro-geochemical environments depending on
parameters such as the extent of decay on arrival at the lagoon floor, in-vivo heavy metal absorption
by the cuticle (Gonzalez-Davila and Millero, 1990), metabolic rates of the microbial mat, localised
sedimentary conditions (physio-chemical), temperature, salinity, degree of anoxia, and pH, among
others. These variables probably explain the range of preservational fabrics found in the insects.
Framboid size varied significantly (1 micron to 25 microns), depending on growth duration (Vietti et

al., 2015). In some parts of the insect, decay was more advanced, resulting in more globular replacement fabrics (Fig. 6: A and B). In other areas, the internal structure of cuticle was replicated, resulting in a mesh-like fabric (Fig. 6: G: A; Barling et al., 2015, fig. 15). Rarely, the pseudomorphing goethite adopts an acicular nanocrystal habit, resulting in 'fuzzy' crystal boundaries at lower magnifications (Fig. 6: H).

In addition to these variations, the model is limited by the current understanding of low temperature pyrite framboid formation (Ohfuji and Rickard, 2005; Hunger and Benning, 2007; Vietti et al., 2015). The precursor protoframboids illustrated in Fig. 11: D are tentatively labelled as iron monosulphides, as this phase is a requirement for pyrite formation (Berner, 1970; Ohfuji and Rickard, 2005). It is unclear if these precursor structures would have been overprinted by pyrite during their formation, or if they persisted for more prolonged periods. The stability ranges (Eh, pH, temperature, etc.) of these initial iron monosulphide phases may have been very small, and they will have only persisted as long as the micro-geochemical environment remained within those ranges. Their persistence is important as their size ultimately controls the size of the subsequent pyrite crystals, which in turn controls the fidelity of preservation. The micro-geochemical environments undoubtedly evolved as continued bacterial sulphate reduction reduced the pH. As a consequence, the fidelity of preservation was controlled by rates of bacterial sulphide reduction. Images of protoframboids recorded by Vietti et al. (2015) suggest that pyrite may first overprint in nano-grains prior to the formation of larger euhedral crystals, but this requires investigation. Here, the overprinting timeframe is 'unknown'.

6. Conclusions

The geochemical processes that allowed for the exceptional preservation of insect tissues in the Crato Formation were dynamic and remain incompletely understood. Nevertheless, recently proposed preservational models have greatly enhanced our understanding of the Crato Formation

578 Lagerstätte. The model presented herein addresses several mismatches between previous models
579 and new observations. The revised model proposes a six-stage process in which three important
580 diagenetic mineral fabrics are precipitated: Fabric 1) framboidal, pseudoframboidal and
581 hemispherical-framboidal precipitation of monosulphide/greigite/pyrite; Fabric 2) nanocrystalline
582 non-framboidal iron monosulphide/greigite replacements, and Fabric 3) 'Orsten'-type calcium
583 phosphate impregnations and encrustations. Although we are unable to demonstrate unequivocally,
584 it is likely that fabric 3 occurs first, as it clearly preserves the most labile tissues. Fabrics 1 and 2 were
585 later pseudomorphed by goethite as a consequence of slow, meteoric weathering of the formation.
586 Calcite infills are also present. Despite the revised model addressing the vast majority of
587 preservation fabrics observed, several rarer fabrics remain unexplained, and are attributed to subtle
588 and localised variations in geochemistry.

589 The 'black' Crato Formation fossil insect from the 'blue' limestone examined does not appear to
590 possess pyrite framboids or pseudoframboids, as would be expected if they indeed represent
591 'unweathered' versions of the same preservational process. Instead, it has a smooth-to-globular
592 surface replacement (see Fig. 5). Here, this is attributed to a mineralised EPS coating over the fossil,
593 obscuring the fabrics below. However, the extremely limited number of 'black' specimens available
594 in this project prevents verification of this suggestion. The idea that these 'black' fossils represent an
595 entirely different preservational process has been eluded to (Osés et al., 2017; Oliveira, 2018), but
596 that interpretation is disagreed with here. This is due to the association of goethite specimens with
597 reduced overburden, which is a strong indicator that their colour difference is a result of meteoric
598 weathering. 'Black' specimens are also informally noted to weather to rusty orange/brown over time
599 if improperly curated (observed by Martill between 2011–2019). Nevertheless, further micron-scale
600 scrutinization of more 'black' specimens is necessary to determine this unequivocally.

601 Further research is now necessary to establish the pre-depositional controls on preservation in the
602 Crato Formation. Although environmental reconstructions have been discussed, only simplistic

descriptions of the processes that transported carcasses to the site of deposition have been presented (i.e. Menon and Martill, 2007). A comprehensive understanding of the palaeoenvironment and ecology of the Crato Formation catchment area(s) is lacking. Once this has been achieved, verification of the model using actuo-palaeontological experimentation can begin. The preservational processes of the Crato Formation are remarkable, preserving animal tissues with sub-micron fidelity. The model herein seeks to explain those processes, but much more research is necessary before we can fully appreciate how these beautiful fossils were formed.

Acknowledgements

We thank Elaine Dyer, Joe Dunlop and Christine Hughes for help with scanning electron microscopy, James Coyne for laboratory assistance and Geoff Long for sample preparation. Ms Florence Gallien is thanked for meticulously preparing specimens and Judith Wohlrabe is thanked for donating such wonderful fossil material. Helmut Tishlinger is thanked for discussions on exceptionally preserved fossils. We thank two anonymous reviewers and Cretaceous Research Editor-in-Chief Eudardo Koutsoukos for their constructive and insightful comments during the writing of this manuscript.

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Figures

Figure 1. Locality map for the Crato Formation. Image adapted from Barling et al. (2015).

809

810 Figure 2. Crato Formation stratigraphic logs and site photographs. A, Individual stratigraphic logs for
 811 the Crato Formation, as outlined in Figure 1. B, Composite stratigraphic log for the Crato Formation.
 812 Logs adapted from Martill and Heimhofer (2007) and Heimhofer et al. (2010). C, Quarry face
 813 exposure of the Nova Olinda Member of the Crato Formation 4 km south-south-west of Nova Olinda.
 814 This exposure displays lightly weathered Nova Olinda Member sediments. D, Fine-scale lamination
 815 typical of the Nova Olinda Member succession.

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Figure 3. A and B, Matrix crystals surrounding insect fossils in the Nova Olinda Member. A, Acid etched Nova Olinda Member sediment sample showing individual rhombohedral calcium carbonate crystals of varying sizes. B, Rhombohedral termini of calcium carbonate matrix crystals preserved in

820 calcite cement moulds, within posterior abdomen of insect. C and D, Examples of the
821 pseudomorphed framboïd (and pseudoframboïd) fabric. C, Arrows highlight subspherical
822 pseudomorphed pseudoframboïds of various sizes, partially fused/inter-grown. D, Arrows highlight
823 pseudomorphed framboïds preserving spiral alignment of microcrystals from original framboïd
824 fabric. E – H, Examples of two different goethite fabrics: the high-fidelity replacement of the
825 external-most epicuticle, and the underlying pseudomorphed framboïd fabric. In each image, *X*
826 highlights exceptionally well-preserved cuticular scales replicated by high-fidelity nanocrystalline
827 impregnation and *Y* highlights globular-to-framboïdal subsurface replacement fabric. Arrows also
828 highlight the sharp contact between the two fabrics, revealing that the high-fidelity replacement is
829 less than 1 µm thick. In H, *Y* shows the ringed structure of a pseudomorphed hemispherical
830 pseudoframboïd. Specimen and photo numbers: A, Specimen UOP-PAL-NB011 (photo 015); B,
831 Specimen UOP-PAL-NB101 (photo 72); C, UOP-PAL-NB022 (photo 30); D, UOP-PAL-NB066 (photo
832 13); E, UOP-PAL-NB111036 (photo 40); F, UOP-PAL-NB111054 (photo 94); G, UOP-PAL-NB111059
833 (photo 46); H, UOP-PAL-NB111036 (photo 66). Scale bars: A, E and G, scale bars = 10 µm; B and C
834 scale bars = 5 µm; D, F and H, scale bars = 1 µm.

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848 Figure 5. Fabric of 'unweathered' black Crato Formation insect cuticle, sputter coated with Carbon.

849 Arrows highlight mostly smooth cuticle, with sub-micron globular aggregates across its surface. No

850 evidence of pseudomorphed framboids (or pseudoframboids) were observed in this specimen.

851 Specimen UOP-PAL-NB111. Scale bar = 2 μm .

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Figure 6. A and B, Examples of loss of original cuticular morphology, associated with perforations in the cuticle. A, X marks area of cuticular fidelity loss, associated with spiracle (or base of disarticulated seta) highlighted by arrow. B, arrows highlight areas of fidelity loss associated with

857 setae. C – F, Examples of internal soft tissues preserved via calcium phosphate impregnation and
858 encrustation ('Orsten'-type preservation). C, Arrows highlight granular fabric along muscle fibre
859 within thorax of winged insect (possibly Hymenoptera), likely flight muscle. D, Diptera limb
860 preserving muscle fibres. Arrows highlight 'Orsten'-type preservation. X highlights calcite mineral
861 infill and Y highlights nanocrystalline replacement (impregnation) of limb cuticle (internal surface). E,
862 arrows highlight muscle fibres in fly genitals (ovary) preserved in calcium phosphate. F, higher
863 magnification image of muscle fibres shown in E, with arrows highlighting the spherical encrustation
864 fabric. G and H, Examples of rare fabrics not represented in the preservational model. G, Areas
865 where internal mesh-work structure has been replicated (highlighted by arrow). This mesh-work is
866 revealed by partial loss of epicuticle (x) adjacent to area where epicuticle is retained (y). H, Examples
867 of nanocrystalline needle-like 'rims' around otherwise high-fidelity replacement of epicuticle,
868 highlighted by arrows. Specimen and photo numbers: A, UOP-PAL-NB036 (photo 37); B, UOP-PAL-
869 NB054 (photo 83); C, UOP-PAL-NB106 (photo 14); D, UOP-PAL-NB104 (photo 42); E, UOP-PAL-NB104
870 (photo 14); F, UOP-PAL-NB104 (photo 18); G, UOP-PAL-NB014 (photo 26); H, UOP-PAL-NB055 (photo
871 92). Scale bars: A, D and G, scale bars = 10 μm ; B and E, scale bars = 50 μm ; C, F and H, scale bars = 1
872 μm .

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875 Figure 7. Energy dispersive X-ray spectroscopic map of specimen UOP-PAL-NB104. A, Sum spectrum.
876 Abundance of gold is due to gold sputter coating. B, Montaged layered energy dispersive X-ray
877 spectroscopic map of specimen UOP-PAL-NB104. Element colour assignments are denoted in the
878 bottom left. Matrix consists of calcitic mineral, fossil cuticle is preserved as an iron oxide mineral,
879 and internal soft-tissues are phosphatic. Remaining images are montaged energy dispersive X-ray
880 spectroscopic map of specific elements: C, Iron; D, Calcium; E, Phosphorus; F, Oxygen. Four-hour
881 spectrum run-time. Kv = 20, WD = 14.5 mm, I Probe = 200 pA. Scale bar = 1 mm.

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Figure 8. Point energy dispersive X-ray spectra of the limb (right forelimb femur) of specimen UOP-PAL-NB104. Spectra 3 and 8 are excluded due to near-identical results to 7 and 1 respectively. Carbon is excluded from this analysis and the abundance of gold is a result of sputter coating. Spectra 1 and 8 reveal the infilling mineral as calcite. Spectra 2 and 4 reveal iron and oxygen preservation of cuticle. Spectrum 5 reveals abundant calcium in the matrix (calcium carbonate). Spectra 6 and 7 reveal calcium phosphate preservation of muscle fibres. Elements marked in red had a 1σ error of $> 10\%$, and are less reliable identifications. kV = 20, WD = 14.5 mm, I Probe = 200 aP. Scale bar = 100 μm .

Figure 9. A, X-ray diffractogram of specimen UOP-PAL-NB114. Blue peaks correspond to calcium carbonate (matrix). Green peaks correspond to goethite. Counts exceed 3,000. Black arrow highlights single unmatched peak, indicating that not all mineralogy is resolved. B, Mineral identifications within specimen UOP-PAL-NB114, along with standard intensity patterns. For the

898 intensity patterns, no vertical measurement is provided as identifications are based on relative
899 intensities and peak position ($^{\circ}2\theta$). Top intensity pattern (orange) is copper (anode material). Middle
900 intensity pattern (blue) is calcium carbonate (Ref. 01-072-4582, scale factor 0.963, $\text{Ca}(\text{CO}_3)$). Bottom
901 intensity pattern (green) is goethite (Ref. 00-008-0097, scale factor 0.165, $\text{Fe}_2\text{O}_3 \bullet \text{H}_2\text{O}$). C, X-ray
902 diffractogram of specimen UOP-PAL-NB111. Blue peaks correspond to calcium carbonate. Green
903 peaks correspond to pyrite. Grey peaks correspond to hydrated calcium sulphate. D, Mineral
904 identifications within specimen UOP-PAL-NB111, along with the standard intensity patterns. For the
905 intensity patterns, no vertical measurement is provided as identifications are based on relative
906 intensities and peak position ($^{\circ}2\theta$). Top intensity pattern (orange) is copper (anode material). Below
907 that (blue) is calcium carbonate (Ref. 01-086-2334, scale factor 0.970, $\text{Ca}(\text{CO}_3)$), followed by (green)
908 pyrite (Ref. 01-079-0617 synthetic, scale factor 0.072, FeS_2), and finally (grey) calcium sulphate
909 [?gypsum] (Ref. 01-070-7008, 0.084, $\text{Ca}(\text{SO}_4) \bullet \text{H}_2\text{O}$).

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Figure 10. Proposed taphonomic pathway for the replacement of goethite Crato Formation insects (part one). **A.** Carcass Arrives: 1. Carbonate sediment. 2. Microbial mat (green) – extracellular polymeric substances. 3. Chains of photosynthetic algae. 4. Setae. 5. Spiracle. 6. Insect internal tissues. 7. Trachea. 8. Internal void. 9. Muscle tissue undergoing minor decay; **B.** Rapid replication of labile tissues in calcium phosphate: 1. ‘Orsten’-type globular preservation of muscle tissues. 2.

917 'Orsten'-type globular encrustation of trachea. 3. Calcite mineral infill in setae void. 4. Calcite infill of
918 void; **C.** Nano-replication of exterior most features in iron sulphides: 1. Sulphate reducing bacteria. 2.
919 Amorphous iron monosulphide replacement of cuticle. 3. In some areas, only the epicuticle is
920 replaced. 4. In other areas, replication penetrates deeper into the epicuticle

921 Figure 11. Proposed taphonomic pathway for the replacement of goethite Crato Formation insects
922 continued (part two). **D.** Coarser replication of remaining internal tissues in iron sulphide
923 protoframoids: 1. Iron monosulphide continues diffuse into the carcass, forming pseudoframboid-
924 precursor structures – obliterating remaining tissue structure. 2. In some areas these intergrew. 3. In
925 others, slightly different conditions resulted in pseudoframboid size. 4. As bacterial sulphate
926 reduction continued, pH lowered; **E.** Stabilisation of iron sulphides by replacement in metastable
927 pyrite: 1. Nanocrystalline pyrite grains overprint iron monosulphide, replicating cuticle with high
928 fidelity. 2. Coarser pyrite overprint on the precursor structures, forming pseudoframboids. 3. Again,
929 these varied in size depending on the conditions; **F.** Oxidation of all iron sulphides to goethite: 1.
930 Millions of years later, the removal of overburden exposes the fossil to oxygenated meteoric water
931 flow. 2. This oxidises the metastable pyrite to form goethite. A written narrative description of these
932 stages is provided in section 5.6. This figure solely addresses the replication of insect tissues. It does
933 not include the mineralisation of EPS around the carcass in calcite or rare siliceous 'halos' around
934 some fossils (see Martill and Barker, 1998; Martill et al., 2007a; Catto et al., 2016).