A cautionary tale about a little-known type of non-nacreous calcareous concretion produced by the *Magilus antiquus* marine snail

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Abstract: Four most unusual objects represented as gastropod-shaped non-nacreous pearls (calcareous concretions) were analysed at the GEMLAB laboratory by standard gemmological and laboratory techniques. They were consistent with natural non-nacreous pearls, but these methods did not provide unambiguous proof that the samples are or are not unusual fossilized gastropods or something else. Therefore the objects were subjected to scanning electron microscopy and finally to a method only rarely used in the gemmological field — ^{14}C age determination. Results were still inconclusive until consultation with malacologists and conchologists led to the conclusion that the specimens are natural and belong to the species *Magilus antiquus*. Their terminology is discussed

Keywords: aragonite, calcareous concretion , calcite, flame pattern, non-nacreous pearl

1. Introduction

Challenges are a daily routine in a gemmological laboratory, but there are times when you are confronted with something that neither you nor your colleagues have ever seen; this paper describes such an event and how it was finally resolved.

Although natural nacreous and non-nacreous pearls are found in a wide variety of different molluscs around the globe, in fresh- and salt-water, due to environmental circumstances natural pearls are becoming increasingly difficult to find from many sources (Strack, 2001). The Philippines is one of the few countries where natural nacreous and non-nacreous pearls are still being found in appreciable but still very limited quantities. In past years the authors have examined many different and exotic natural pearls from this and other locations, but the four gastropod-shaped objects described as pearls and submitted to the GEMLAB for identification were by far the most unusual to date.

It is known that objects or animals trapped within a mollusc may end up as blisters or even as blister pearls, since the mollusc reacts to the intrusion of an object/animal with the formation of nacre (Strack, 2001). Therefore worms, small bivalves and even fishes can be found within blister pearls. Such intruder-formed blisters are often at least partially hollow and the object/animal can always be found in the interior (Hainschwang *et al.,* 2009).

If the gastropod-shaped objects described in this paper are pearls that would indicate that the captured gastropods had not been covered by $CaCO_3$, but that the hollow shells of the animals had been completely filled by the pearl substance. The difficulty of explaining the formation of such pearls and their unusual shapes raised doubts about their identity and the final task was to investigate whether they were counterfeits, fossilized gastropods or some other type of material.

2. Materials and Methods

Figure 1: The largest gastropod-shaped sample of 84.77 ct, represented as a natural non-nacreous white pearl. Photo by T. Hainschwang.

Four non-nacreous white gastropod-shaped specimens weighing 7.06, 14.99, 19.74 and 84.77 ct *(Figure 1)* were studied.

They were examined using light microscopy, fluorescence microscopy, scanning electron microscopy, ultraviolet visible near infrared (UV-Vis-NIR) spectroscopy, Fourier transform infrared (FTIR) spectroscopy, photoluminescence spectroscopy, energy dispersive X-ray fluorescence (EDXRF) and scanning electron microscope/energy dispersive X-ray (SEM-EDX), chemical analysis, radiography and finally ^{14}C age determination.

Microscope observations were made in reflected light and darkfield illumination using a Leica M165C binocular microscope equipped with a Schott LED light source and a Leica DFC420 CCD microscope camera with a resolution of 5 megapixels.

Luminescence of the samples was observed using a standard long wave and shortwave ultraviolet radiation lamp (365 and 254 nm respectively) and by the prototype of the GEMLAB fluorescence microscope using a selectively filtered 200 W xenon light source adjustable to a tuneable monochromatic excitation light source whenever necessary.

SEM imaging and semi-quantitative chemical analysis were carried out using a Zeiss Supra 40VP scanning electron microscope equipped with an EDAX Genesis 2000 EDX spectrometer at the research facilities of Ivoclar in Schaan/Liechtenstein.

Specular reflectance infrared spectra of all specimens were recorded in the range of 4000–400 cm^{-1} at 4 cm^{-1} resolution with a Perkin Elmer Spectrum BXII FTIR spectrometer. The instrument was equipped with a DTGS detector and a KBr beam splitter. A Perkin Elmer fixed angle specular reflectance accessory was used for experiments.

Reflectance UV-visible-near infrared absorption spectra in the 240–1050 nm range were recorded for two samples with the prototype of the GEMLAB Xenon UV-Vis-NIR spectrometer system with a resolution of 0.6 nm; the samples were examined in a reflectance setup within a custom made integration sphere.

Photoluminescence (PL) spectra of two specimens were recorded using a custom-built system using 532 nm and 473 nm diode-pumped solid state lasers coupled to an Ocean Optics Maya spectrometer with a resolution of 0.9 nm.

Quantitative EDXRF chemical analysis of all samples was carried out with a custom-built system equipped with a 40 kV X-ray tube and a thermoelectrically cooled Si detector; the analyses were done in air and for one sample under a helium atmosphere. Quantitative chemical analysis of one sample was carried out using the EDX system of the scanning electron microscope described above.

A ^{14}C isotope analysis of one specimen was carried out at the accelerator mass spectrometry (AMS) facilities of the Ion Beam Physics Department, ETH Zurich, for age determination. For this destructive method a small piece weighing 0.015 g was carefully removed from one of the objects and dissolved in concentrated phosphoric acid. The released CO_2 was then graphitized (Hajdas *et al.,* 2004) and pressed into cathodes for AMS measurements using a mini radiocarbon dating system (MICADAS) (Synal, 2007). In this method ions obtained from the graphite are accelerated to high kinetic energies to enable separation of the ^{14}C and ^{12}C isotopes.

For comparison, samples of aragonite (crystals, stalactitic aggregates ['eisenbluete']), calcite crystals and natural nacreous and non-nacreous pearls were analysed by FTIR, UV-Vis-NIR and photoluminescence spectroscopy as well as EDXRF chemical analysis.

3. Results and discussion

3.1. Visual observation, microscopy and luminescence

The four objects in this study apparently belong to the same species of gastropod; their form and shape are similar and there is variation in only minor details *(Figure 2)*. The samples were all non-nacreous white, semi-translucent, and their surfaces show great details of the original shell without any indications whatsoever of polishing or any other form of working, except that the original aperture of the gastropods showed indications that they were polished/worked.

All four specimens contain small holes such as that in the 7.06 ct specimen (*Figure 3a*); such holes are present in most sea shells and can be caused by drill molluscs or parasitic worms. Microscopic observation showed a distinct concentric structure in the zone of the original aperture of the shell, and in the 7.06 ct specimen there is a central grain surrounded by darker matter (*Figure 3b*).

A faint flame pattern is present in all samples but only clearly visible when the surface of the pearls was viewed carefully under the microscope *(Figure 4)*. Such a flame pattern is characteristic for certain non-nacreous pearls, of which the pink to orange pearls from Strombus gigas and Melo melo gastropods are the best known (Hänni, 2009; Strack, 2001). Besides

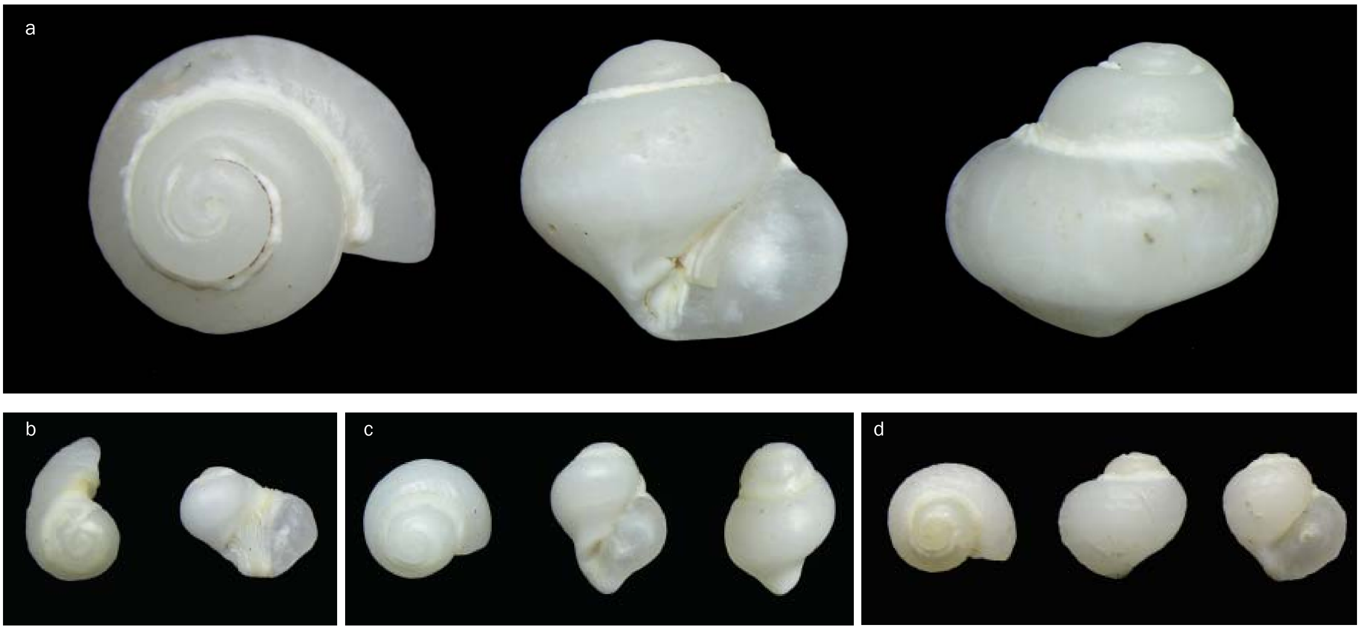


Figure 2: Different views of the four specimens examined in this study: (a) 84.77 ct, (b) 19.74 ct, (c) 14.99 ct and (d) 7.06 ct. The general shape of the specimens indicates that they belong to the same or a very similar species of gastropod. Photos by T. Hainschwang.

these pearls, this kind of flame pattern is also known from other non-nacreous gastropods and certain bivalve molluscs.

Under intense UV illumination, as used in the GEMLAB fluorescence microscope, all specimens exhibited a distinct blue fluorescence, reminiscent of the emission known for many pearls. In three specimens a lighter and yellower fluorescence was visible in the central portion of the gastropods' original aperture *(Figure 5)*. This emission followed the most evident concentric structures visible.

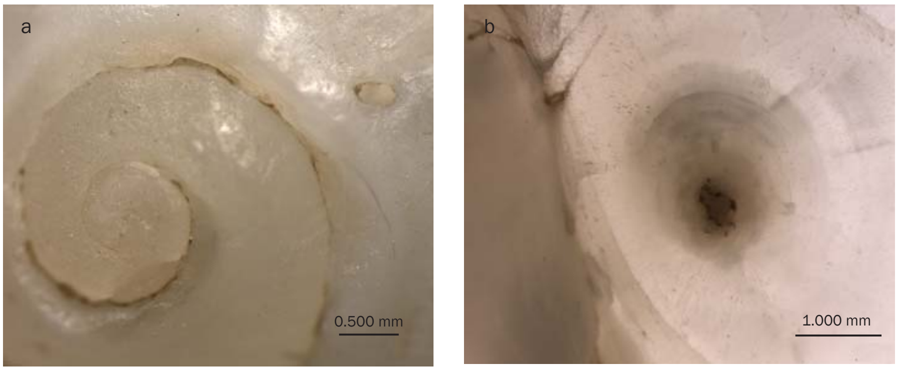


Figure 3: Close-up views of the 7.06 ct specimen: (a) view of the spiral and of a parasite-caused drill hole; (b) the concentric structure and central grain at the original aperture of the gastropod. Photos by T. Hainschwang.

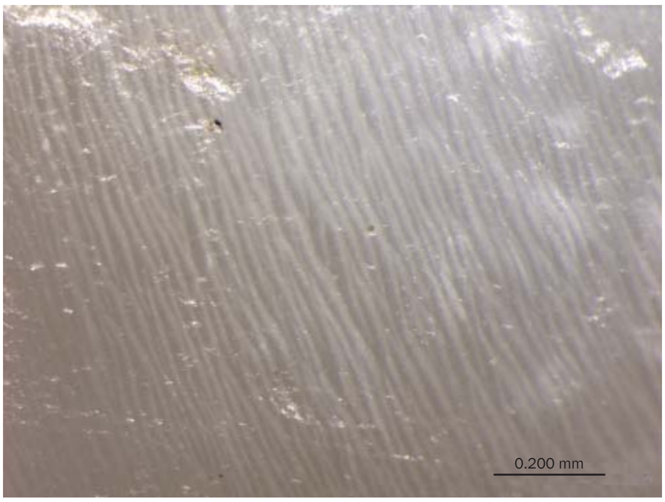


Figure 4: The fine flame pattern seen in the largest specimens, as seen in reflected light. Photo by T. Hainschwang.

Figure 5: The 7.06 ct gastropod-shaped specimen under strong UV excitation shows distinct zoning at the gastropod's original aperture. Photo by T. Hainschwang.

A little-known type of non-nacreous calcareous concretion produced by the Magilus antiquus marine snail

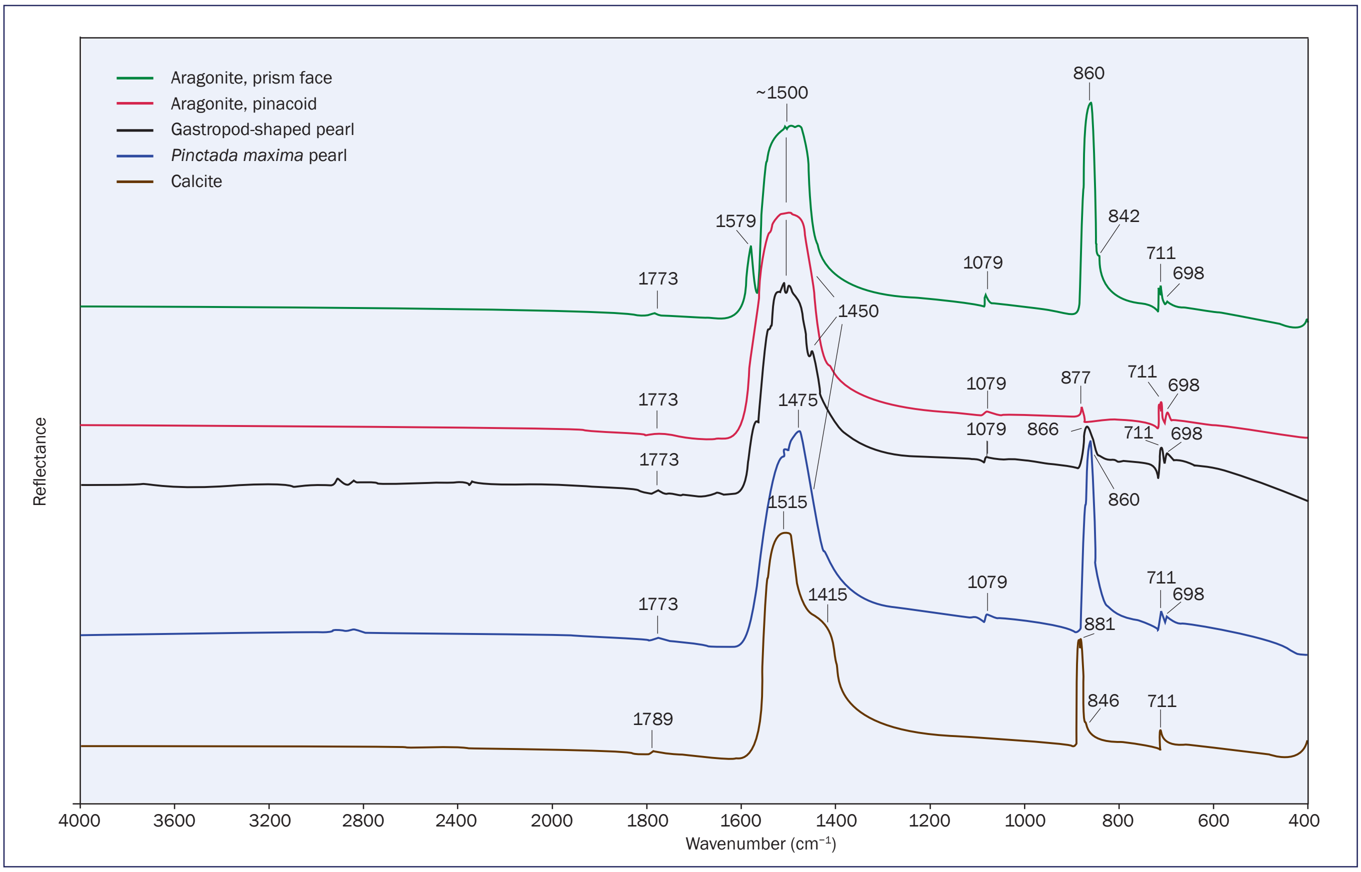


Figure 6: Specular reflectance infrared spectra of aragonite, calcite, a non-nacreous white pearl and a gastropod-shaped sample.

The initial results from these classical gemmological approaches seemed to indicate that the gastropod-shaped specimens were neither forgeries nor fossils; at the time of analysis in the laboratory it was believed that non-nacreous pearls and certain sea shells were the only materials exhibiting the characteristic flame structure visible in *Figure 4*. The lack of banded layers in strong transmitted light indicated that the objects could not be polished from shell, which is a common type of forgery to imitate non–-nacreous natural pearls. The specimens appeared to be most unusual pearls, but the fact of the existence of such gastropod-shaped pearls would contradict all classical explanations of pearl formation and so further examination to try and clarify how they were formed was desirable.

3.2. EDXRF chemical analysis

Using EDXRF analyses major calcium and minor but distinct strontium were detected, a perfect match with what would be expected when testing saltwater aragonitic materials. The near-absence of manganese is consistent with a saltwater origin (Hänni *et al.,* 2005). One sample (7.06 ct) was additionally analysed by SEM-EDX, with identical results. For comparison, EDXRF data were collected for aragonite (crystals, stalactitic aggregates ['eisenbluete']), calcite crystals and natural nacreous and non-nacreous pearls. The chemical compositions of the nacreous and non-nacreous pearls were practically identical, while the aragonite and calcite mineral samples all contained significantly lower strontium.

3.3. Specular reflectance FTIR spectroscopy

Specular reflectance FTIR spectra recorded from all specimens confirmed that their main constituent is aragonite. The commonest polymorphs of $CaCO_3$ — calcite and aragonite — can be easily distinguished using this method since their band positions vary slightly and aragonite also exhibits weak features around 1085 cm^{-1} and 697 cm^{-1} which are absent in the calcite spectra *(Figure 6)* (Hainschwang and Notari, 2008). Certain non-nacreous pearls are calcitic (e.g. Pinna pearls), but we have not yet had a white non-nacreous pearl that was dominantly calcitic in the GEMLAB laboratory; so far all have been dominantly of aragonitic composition.

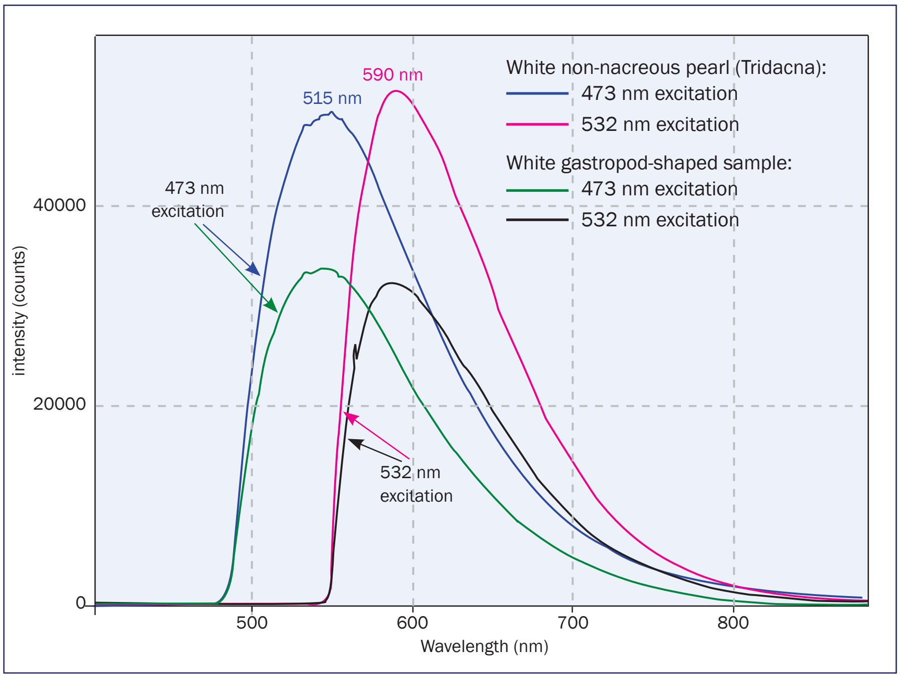
3.4. Reflectance UV-Vis-NIR spectroscopy

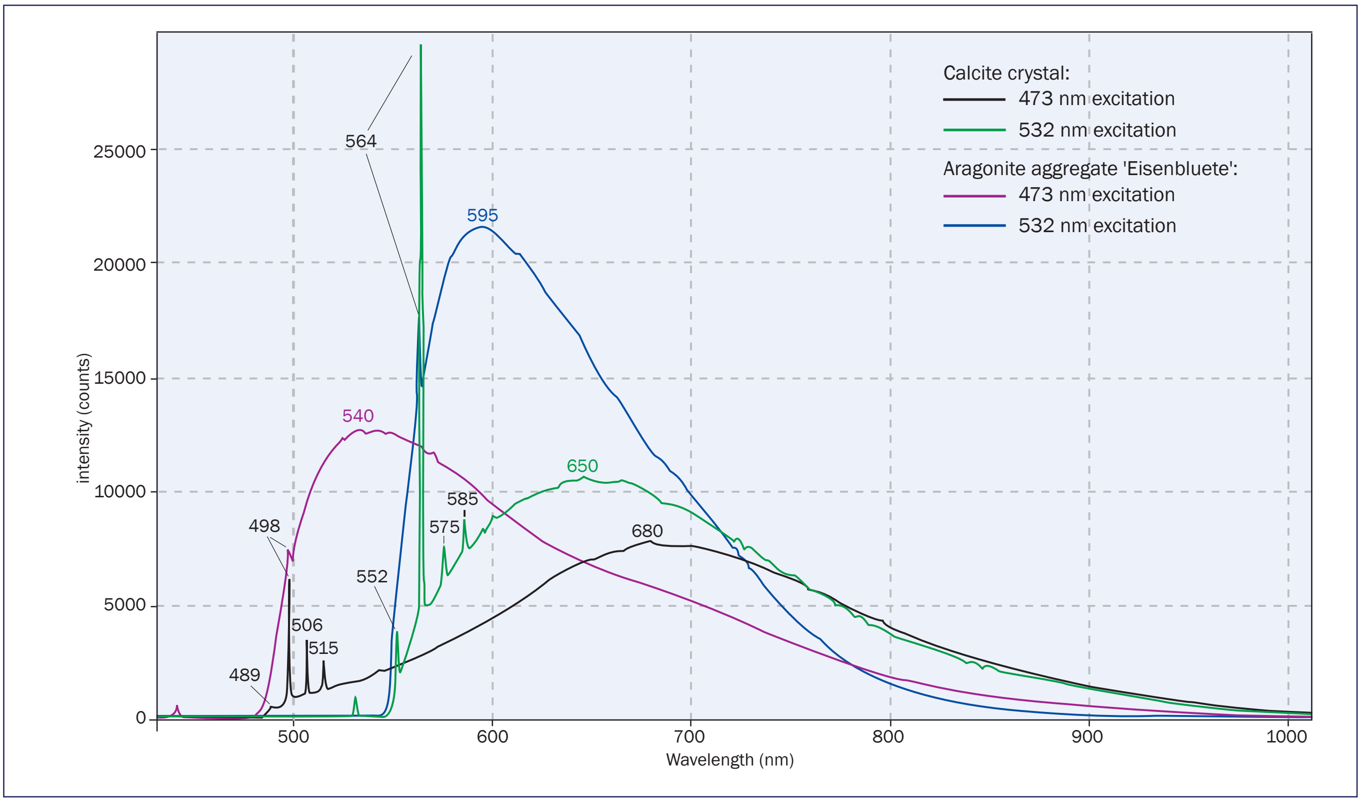
The spectra of all four specimens are identical to the spectra of white nacreous and non-nacreous pearls, with a strong absorption band at 285 nm; this band is characteristic for all aragonitic materials, including aragonite itself and is thus of no diagnostic value to distinguish a white non-nacreous pearl from other aragonitic materials.

3.5. PL and Raman spectroscopy

The PL spectra recorded with 473 nm

Figure 7: The PL spectra excited with 532 and 473 nm lasers of a non-nacreous white pearl and a gastropod-shaped sample (left), compared to aragonite ('eisenbluete') and calcite (right). The sharp features present and indexed in the graphs are Raman scattering peaks.





and 532 nm laser excitation are identical to the spectra obtained from non-nacreous white pearls. The spectra consist of a simple broad band emission centred at about 545 and 590 nm respectively with sharp peaks corresponding to the Raman lines of aragonite. The fibrous aragonite 'eisenbluete' samples that were used for comparison exhibit a very similar broad band PL while calcite shows a different broad band emission centred at 680 nm (473 nm excitation) and 650 nm (532 nm excitation) respectively *(Figure 7)*.

3.6. Radiography

Radiography of the samples revealed that the objects are all solid. Little structure is visible and no remnants of, for example, the original shell that could be trapped within the objects were found. The only significant features are the drill holes that can also be seen under the microscope *(Figure 8)*, and some of these holes cross the whole specimen. The

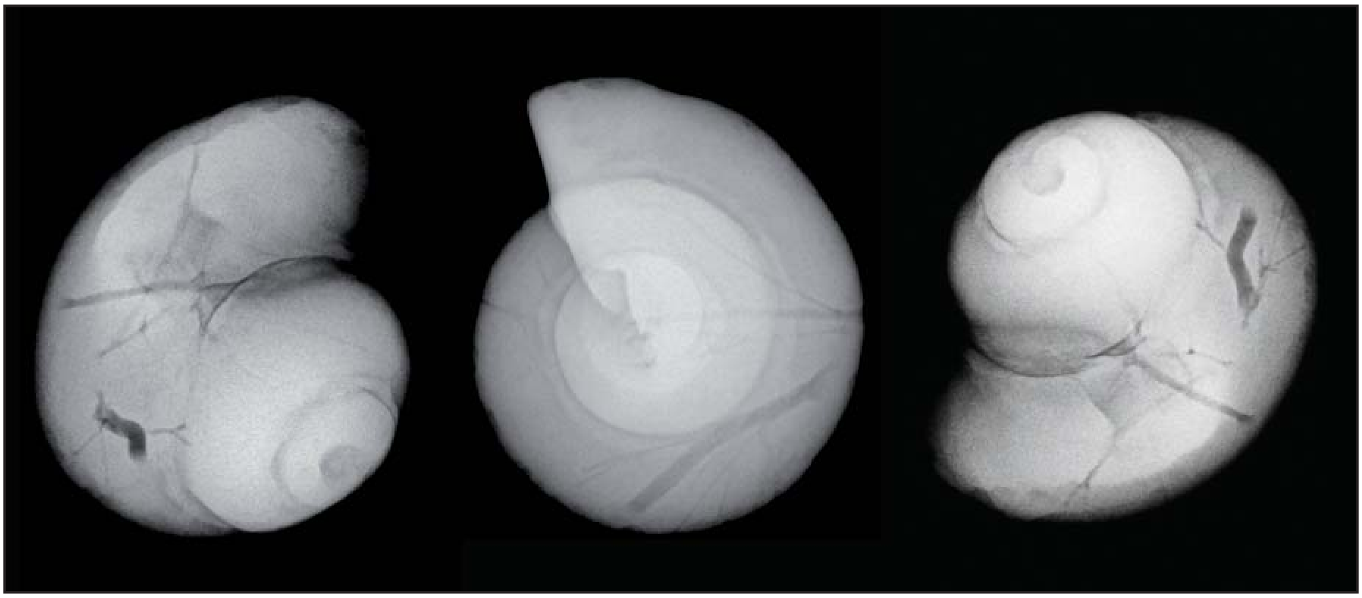


Figure 8: Radiographs in three different orientations of the largest gastropod-shaped sample. The grey linear features of variable width represent the hollow drill channels present in all the specimens.

extremely subtle structure of natural non-nacreous white pearls is — in the authors' experience — generally not visible in conventional radiography. When white non-nacreous pearls are sawn in half, there is normally no evidence of circular growth structure and only diffuse bands of slightly variable coloration can be seen.

The radiographs clearly pointed towards naturally grown solid pearl-like objects.

3.7. SEM

The 7.06 ct specimen was examined using the SEM in order to get a better picture of the structure of the material *(Figure 9)*.

The SEM images revealed platy and fibrous crystals of aragonite, which were in part arranged in a concentric structure. The fibres are not all near-parallel but are distributed in the form of 'bundles', some of which have a V-like appearance. The presence and distribution of these fibres is the reason for the flame pattern that is visible on the surfaces of the specimens (Hänni, 2009). These results are consistent with what would be expected from a natural non-nacreous pearl.

Although the presence of the concentric structure at the original aperture indicated that some of the original piece may be missing, there was no evidence of polishing; a possibility would be that part of the sample was broken off the original piece.

3.8. ^{14}C age determination

Radiocarbon dating was carried out to determine whether or not the specimens could be an uncommon type of fossilized sea mollusc.

The analysis of a small piece removed from one specimen indicates that the material was no more than 400 to 500 years old. That is, its age is well within the historical period and therefore it is not a fossil. Also, this is consistent with the fact that most aragonite transforms to calcite when aragonitic shells are fossilized. Aragonite in fossils is only known in the iridescent surface layers of so-called 'ammolite'.

4. Discussion

The data obtained using the above methods prove that the samples are not man-made forgeries, that they have not been polished from shell material and that they are not fossils. The composition of the specimens was identified as principally aragonite with distinct traces of strontium, radiography proved their solid nature and the presence of concentric growth structure combined with a very fine flame pattern due to fibrous aragonite growth in all samples, clearly points to the specimens being natural non-nacreous pearls.

Because we had no plausible explanation for the formation of solid non-nacreous gastropod-shaped pearls, we sought an expert in molluscs and shells (a malacologist and conchologist) who could identify the species that was represented by these specimens. The information obtained from two conchologists was a revelation: the specimens were confirmed to be of natural origin, but they were not formed inside another marine mollusc, but by the gastropod itself (Massin, 1982). There are some most unusual gastropods that live on and in, and feed from corals ('coral dwellers') and that grow together with the coral. Our four specimens were identified as *Magilus antiquus* (Montfort, 1810), belonging to the family Muricidae (subfamily Coralliophilinae, a name meaning 'liking coral'). When the coral grows, the *Magilus* fills up its shell with aragonite and lives on some sort of pedestal close to the surface of the coral (Oliverio, 2009, Figs 11F, G). Complete specimens usually have a rather long tube-like uncoiled shell; this solid tube-like 'prolongation' of the juvenile coiled shell is used as a 'house' by the animal, which can live close to the surface of the

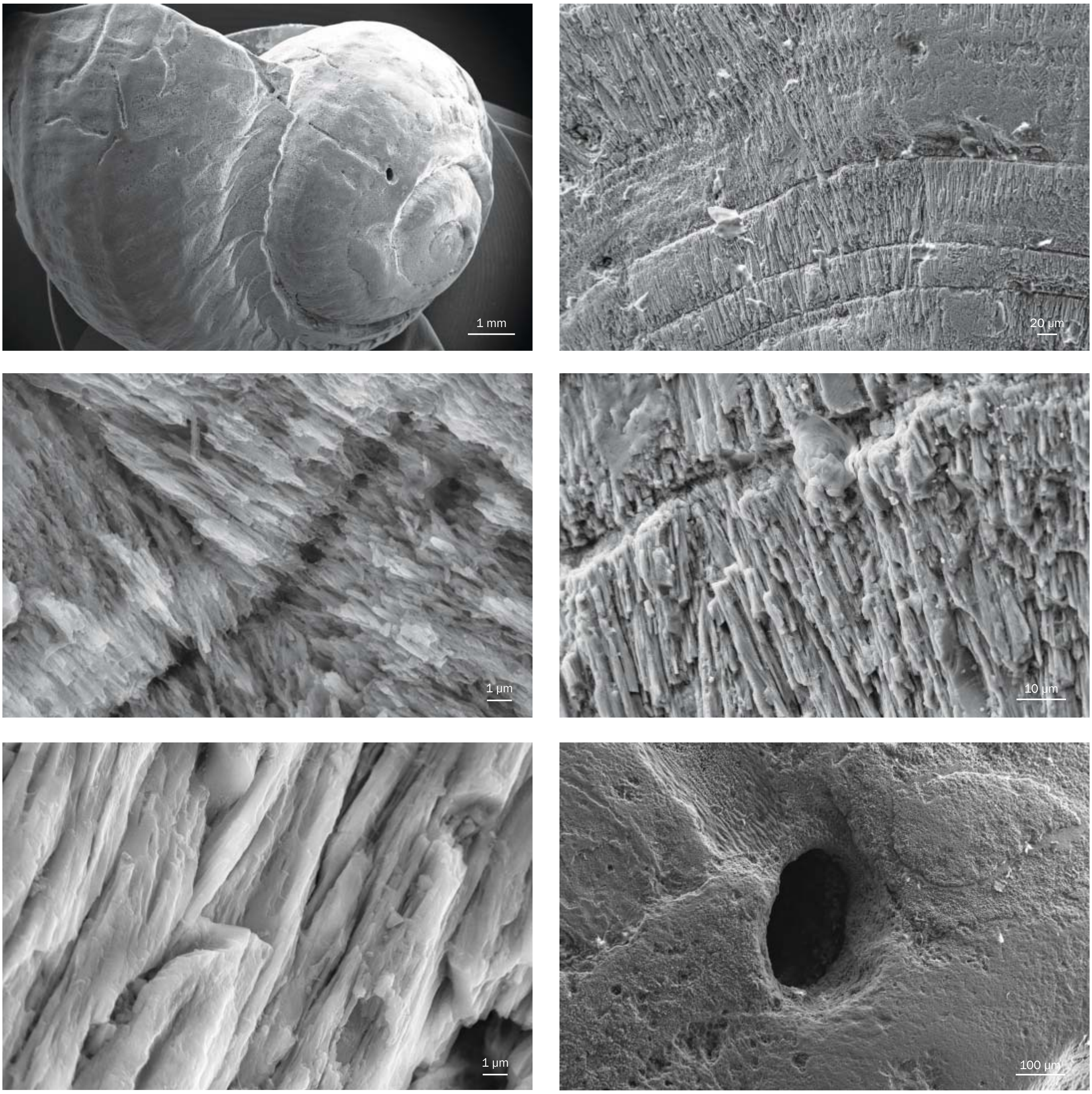


Figure 9: SEM images showing details of the gastropod-shaped specimen of 7.06 ct.

coral thanks to the pedestal. As the coral grows, the animal produces more and more aragonite to keep the pedestal at about the same distance from the coral surface. The shell of this gastropod is as white as the 'filled' pieces described in this study, but with time it becomes absorbed in the juvenile part. In the parts of the gastropod buried deeply in the coral, there is rarely any trace of the original shell remaining.

5. Conclusions

The four specimens described above constitute a clear demonstration that the utmost care must be taken before drawing conclusions about uncommon or new materials. The specimens of *Magilus antiquus* were sold as natural non-nacreous pearls and this is the basis on which they were examined. In this outstanding case the material showed exactly the same properties as the material that it was meant to imitate (a natural non-nacreous pearl); only the fact that there was no explanation for the formation of these objects finally led to the discovery of the real nature of these 'pearls'.

Concerning the nomenclature of such calcareous concretions formed by coral-dwelling gastropods, it is unclear whether they should be declared as 'non-nacreous pearls' or simply as 'calcareous concretions'; probably the material, which demonstrably represents the filled shell of a mollusc, should be called a calcareous concretion. Although it is formed by marine gastropods, the process and motivation of the formation is quite different from that of 'regular' non-nacreous pearls and the fact that the animal lives on this formed material makes the decision on what to call these 'magilus pearls' an ambiguous one.

Despite the doubts over their name, these unusual objects could make attractive and very individual pieces when mounted in jewellery.

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