Article

Cyanobacterial fossils in terminal Paleoproterozoic ferric microbialites (Xuanlong type iron formation) in Hebei, China

中国河北古元古代末长城系串岭沟组铁质微生物岩宣龙式铁矿中发现丝状蓝细菌胶鞘化石

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Abstract

The stratiform iron formation in the terminal Paleoproterozoic Chuanlinggou Formation (1650-1640 Ma) in Xuanhua-Longguan area of Hebei Province, China, i.e., the Xuanlong-type iron formation, is one of the important iron ore types in China. Some researchers considered it had a microbial origin, but did not give convincing evidence. In this study, rich filamentous fossils were found from an iron bed. The filaments are 3.9~26.7 μm in diameter. Based on their shape and size, the fossils are identified as sheath fossils of cyanobacteria. The presence of the fossils indicates that the Xuanlong-type iron formation formed in the marine euphotic zone, and cyanobacteria may have provided oxygen for the formation of the iron ore.

Key words: Xuanlong-type iron formation; iron formation; iron oolite; ferric ooid; iron stromatolite; ferric oolite; ferric stromatolite; sheath fossil; cyanobacteria; Paleoproterozoic; Changchengian System; Chuanlinggou Formation

1. Introduction

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A several meters thick iron formation occurs in the bottom of the Paleoproterozoic Changchengian System Chuanlinggou Formation (1.65-1.64 billion years ago) in the Xuanhua-Longguan-Chengde area, Hebei Province, China. In the 1920s, Swedish geologist Andersson (1924) named it Xuanlong-type iron formation according to its regional distribution, and considered it a shallow marine chemical deposit. Since the 1970s, Chinese geologists have done a lot of research on the iron formation, and proposed that the formation of the Xuanlong-type iron formation is the result of cyanobacteria and other microbial biomineralization (Meng, 1979; Zhu, 1980; Chen, 1982; Hou et al., 1983; Liu, 1993; Zhao, 1994; Liu et al., 1995; Liu et al., 1997; Dai et al., 2003; Lin et al., 2019).

Microbiological fossils were reported from the Xuanlong-type iron formation (Du et al., 1999; Dai et al., 2003; Lin et al.,2019) and were considered key evidence of microbial contribution in formation of the Xuanlong-type iron formation. The presence of fossils can provide important information about the formation environments of the Xuanlong-type iron formation. However, the fossils previously reported are not typical and need to be confirmed. The purpose of this study is to look for microbial fossils from the iron formation, to determine if they are important in the formation of the iron formation. After examination of 11 thin sections with microscope and 8 rock samples with ESM, rich filamentous microbial fossils were found in one sample, and their systematic affinities were identified based on their shape and size. The existence of cyanobacterial fossils indicates that cyanobacteria may have played an important role in the formation of the Xuanlong type iron formation.

2. Location and stratigraphy

The Xuanlong-type iron formation is distributed in the Xuanlong syncline (Xuanhua Depression) in the northwest Yanshan trough belt of the North China paraplatform (Fig. 1 (a), (b)), and is mainly distributed in the Zhangjiakou, Chengde, Baoding and Shijiazhuang areas, Hebei Province, China. The strata in this area include the Neoarchean Sanggan Group, the Paleoproterozoic Changchengian Sys-

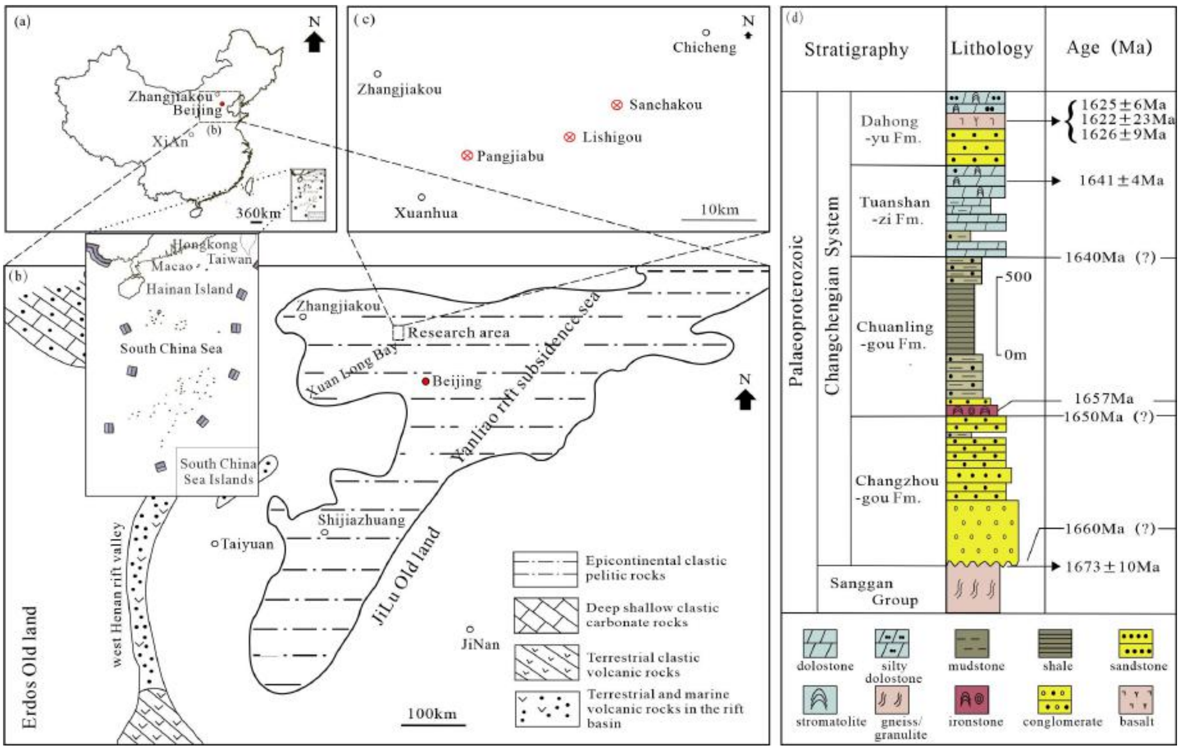
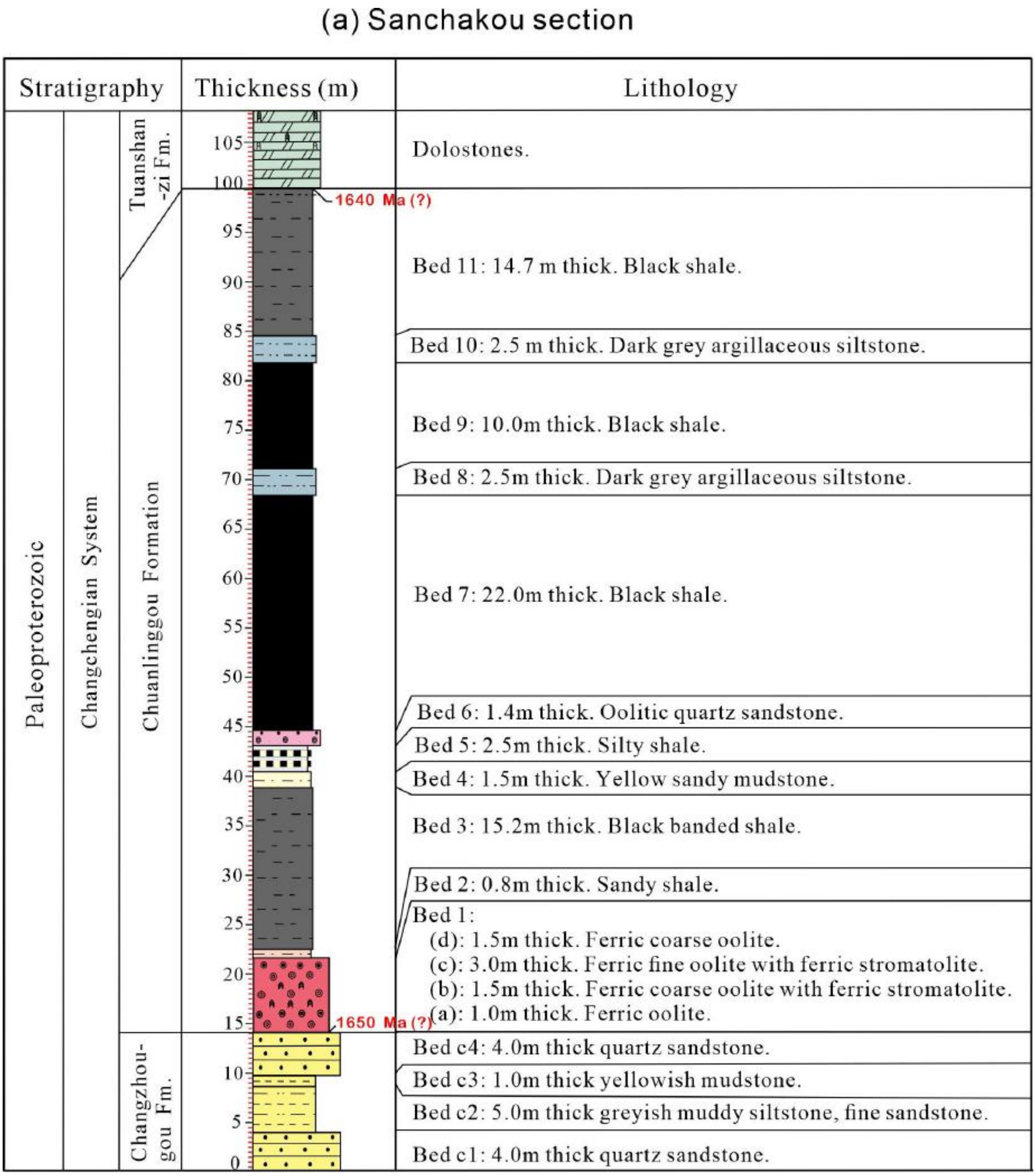
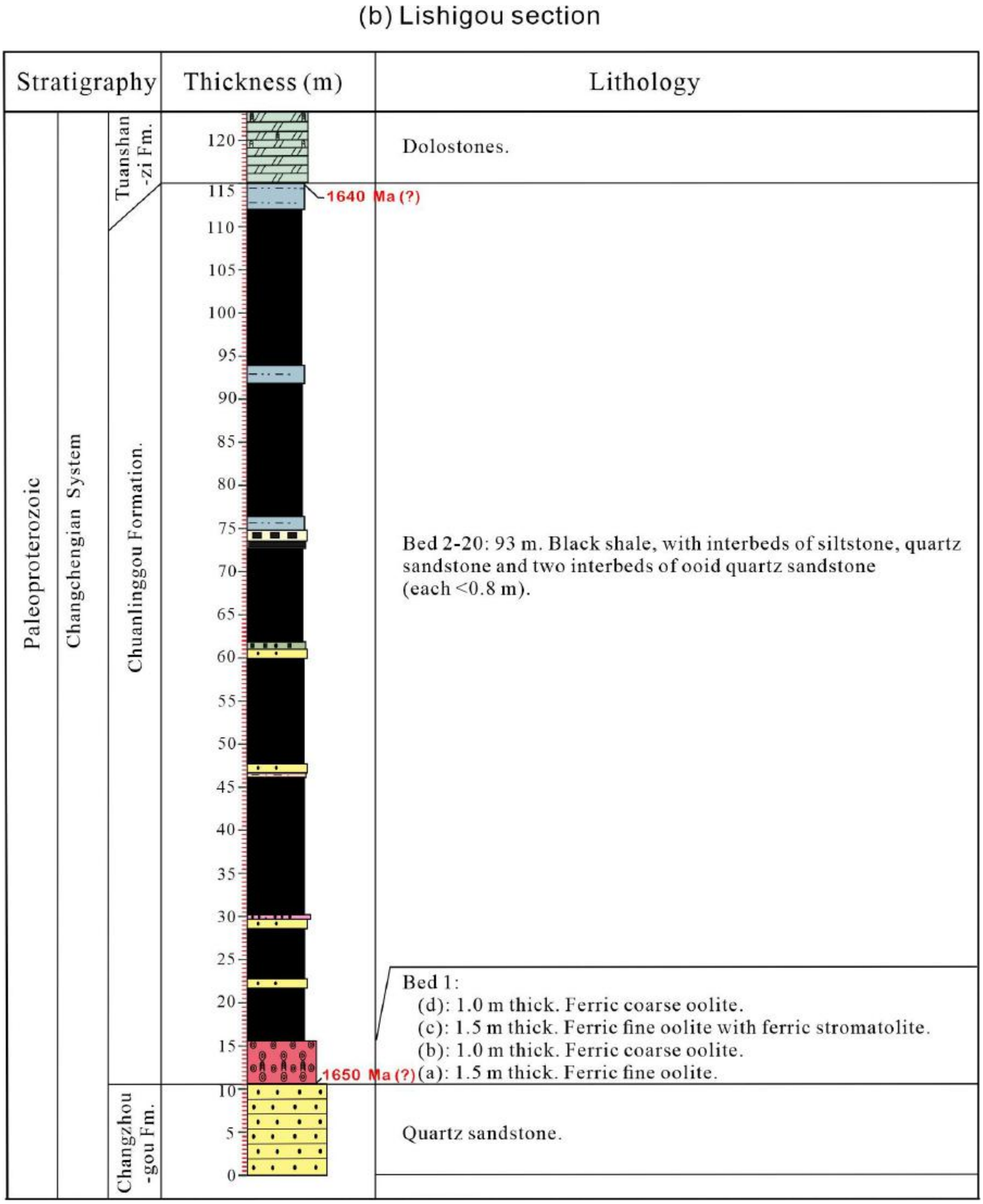


Fig. 1 Location and stratigraphy of the Xuanlong-type iron bed at the base of the terminal Paleoproterozoic Changchengian System Chuanlinggou Formation in the Xuanhua-Longguan area, Hebei Province, China (modified from Wang et al., 1985；Lu et al., 1991, 2008；Gao et al., 2008；Zhang et al., 2013；Li et al., 2013；Duan et al., 2018; Lin et al., 2019).



tem, the Mesoproterozoic Jixianian System and the unnamed system, the Neoproterozoic Qingbaikou System, the Mesozoic Jurassic System, and the Cenozoic Neogene System (Du et al., 1999). The Changchengian and Jixianian systems are developed in this region. The Changchengian System can be divided into the Changzhougou Formation, Chuanlinggou Formation, Tuanshanzi Formation and Da-



hongyu Formation (Fig. 1 (d)) from the bottom up, and is mainly composed of clastic rocks, argillaceous (silty) rocks and carbonate rocks. The Xuanlong-type iron formation occurs at the base of the Chuanlinggou Formation.

The Chuanlinggou Formation in the study area is in conformable contact with the dolostone of the overlying Tuanshanzi Formation and the quartz sandstone of the underlying Changzhougou Formation. The Chuanlinggou Formation is mainly composed of black shale, argillaceous siltstone, sandy mudstone and oolitic quartz sandstone, and the 5-7 m thick Xuanlong-type iron formation in its base. The

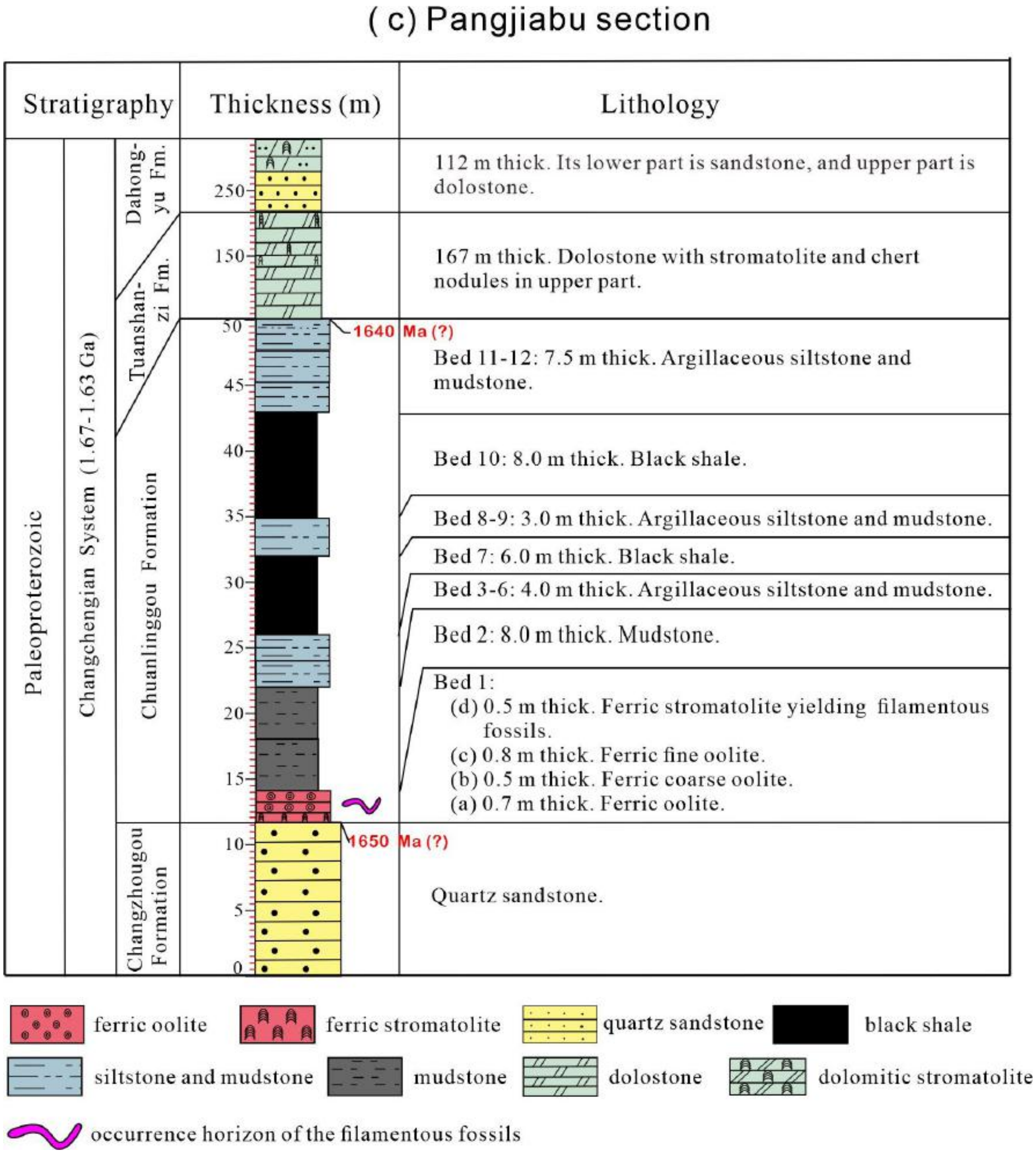


Fig. 2 Stratigraphy and lithology of the terminal Paleoproterozoic Changchengian System Chuanlinggou Formation of the three stratigraphic sections at Sanchakou, Lishigou and Pangjiabu in Xuanhua-Longguan area, Hebei Province, northern China (modified from Lin et al., 2019).

iron formation consists of 1 to 4 beds of ferric stromatolites and ferric oolites. The Changzhougou Formation is mainly composed of quartz sandstone, argillaceous siltstone and fine sandstone. The Tuanshanzi Formation is mainly composed of dolomitic stromatolite.

In this study, three typical stratigraphic sections with Xuanlong-type iron formation in Xuanhua-Longguan area, including Sanchakou (40°48 '05.01 "N, 115°40' 27.76" E), Lishigou (40°74 '9.57 "N, 115°63 '93.02 "E) and Pangjiabu (40°48' 05.01" N, 115°40 '27.76 "E) (Fig. 2), were examined, measured and sampled. In the three sections, the iron formation occurs at the base of the Chuanlinggou Formation, and generally consists of four beds ((a), (b), (c) and (d) of Bed 1 in Fig. 2).

3. Materials and methods

Measurement and sampling on the outcrops of the Pangjiabu section were carried out. More than 100 hand specimens of silver-gray ferric oolites and ferric stromatolites were collected from Sanchakou, Lishigou and Pangjiabu sections.

A total of 39 thin sections were made from the iron formation samples, and examined with microscope Olympus BX41-P at Institute of Geology and Geophysics, Chinese Academy of Sciences (IGGCAS). Seven samples of 2 × 3 × 0.3 cm size for SEM observation were taken from 5 iron formation samples, including 5 samples of ferric stromatolites (SCK-14, from the Sanchakou mine; PJBkc8 from the Pangjiabu section; PJBhfz9-a-1, PJBhfz9-a-2, and PJBhfz9-b-1 from the Pangjiabu mine) and 2 samples of ferric oolites (PJB33 from the Gate 33 of the Pangjiabu mining tunnel and SCK1 from the Sanchakou mining tunnel), coated with carbon in vacuum chamber, and observed with SEM (ZeissGemini450)-Raman (WITecAlpha300R) and Focused Ion Beam-Scanning Electron Microscope (FIB/SEM) systems (Zeiss Auriga Compact) online at IGGCAS.

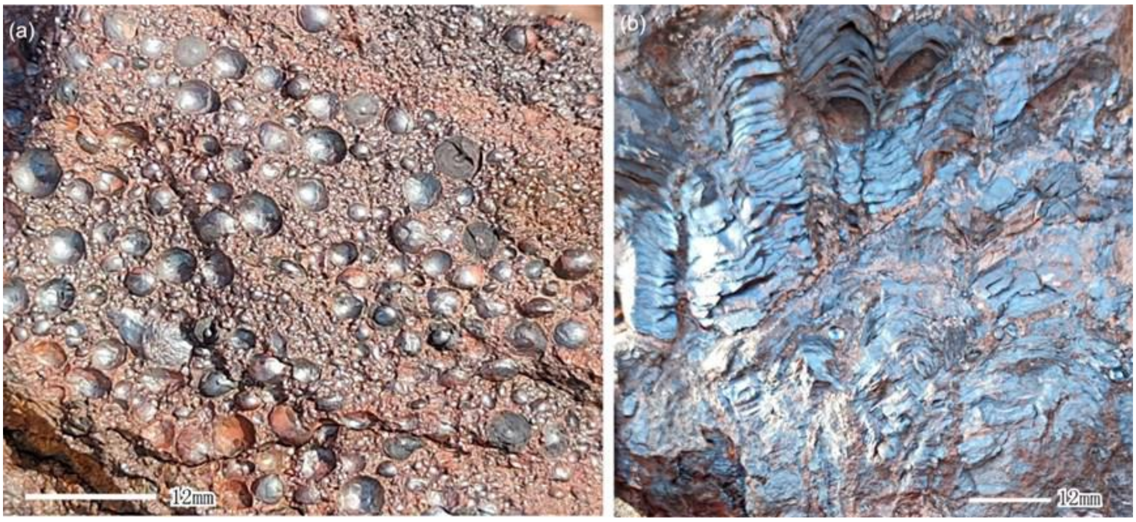


Fig. 3 Hand samples of ferric oolite (a) and ferric stromatolite (b) from the Xuanlong-type iron formation at the base of the terminal Paleoproterozoic Chuanlinggou Formation in Xuanhua-Longguan area, Hebei Province, China. The diameter of the ooids ranges 1-3 mm.

4. Filamentous fossils from the ferric stromatolite

4.1 General features of the ferric oolites and ferric stromatolites

According to the results of Raman spectral analysis (Fig. 4 (c), (d)) and energy spectrum analysis, the mineral composition of the studied specimens is mainly hematite, with a small amount of siderite, limonite, pyrite and quartz debris. The iron formation mainly consists of ferric oolites and ferric stromatolites. The ferric ooids are spherical and elliptical (Fig. 4 (a)), with a diameter ranging about 0.1-0.3 cm, and are composed of very regular concentric hematite laminae. Observation with SEM shows that the laminae consisted of flaky hematite minerals (Fig. 4 (e), (f)). The ferric stromatolites are in silver color, approximately cylindrical, generally about 0.5 to 1.0 cm in diameter (Fig. 4 (b)) and are composed of very regular upper-arching laminae. The flaky hematite crystals of the laminae in the ooids and stromatolites are 1-5 μm in size (Fig. 4).

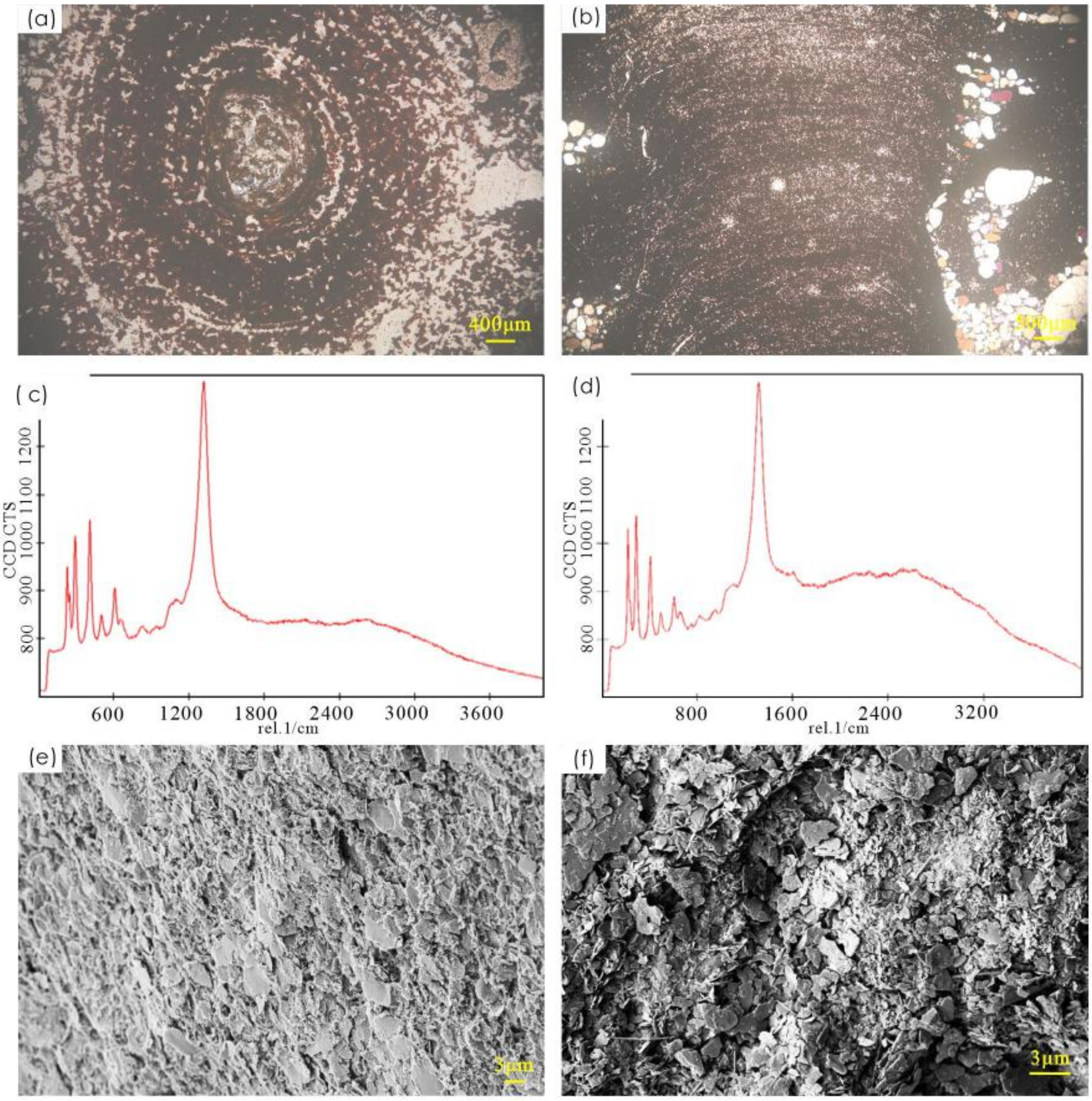


Fig. 4 Microscopic features of the ferric oolites and ferric stromatolites in the terminal Paleoproterozoic Chuanlinggou Formation in Xuanhua-Longguan area, Hebei province, China.
(a) and (b) are photomicrographs taken with Olympus BX41. (a): a ferric ooid with obvious laminated fabric. (b): a ferric stromatolite with obvious laminated fabric. (c) and (d): Raman spectrum photos of the laminae of a ferric ooid showing a strong peak at about 1300(<1330) indicating it is hematite. (e): laminae of a ferric ooid consisting of flaky hematite crystals. (f): laminae of a ferric stromatolite consisting of flaky hematite crystals.

4.2 Morphological characteristics of the fossils

Filamentous fossils were found in PJBhfz9-a-1, a ferric stromatolite sample from Pangjiabu section. Total 13 filaments were exposed in the breaking surface of the sample. They all have one end or a part embedded in the sample, indicating that they are not contaminants. Some filaments occur on other fila-

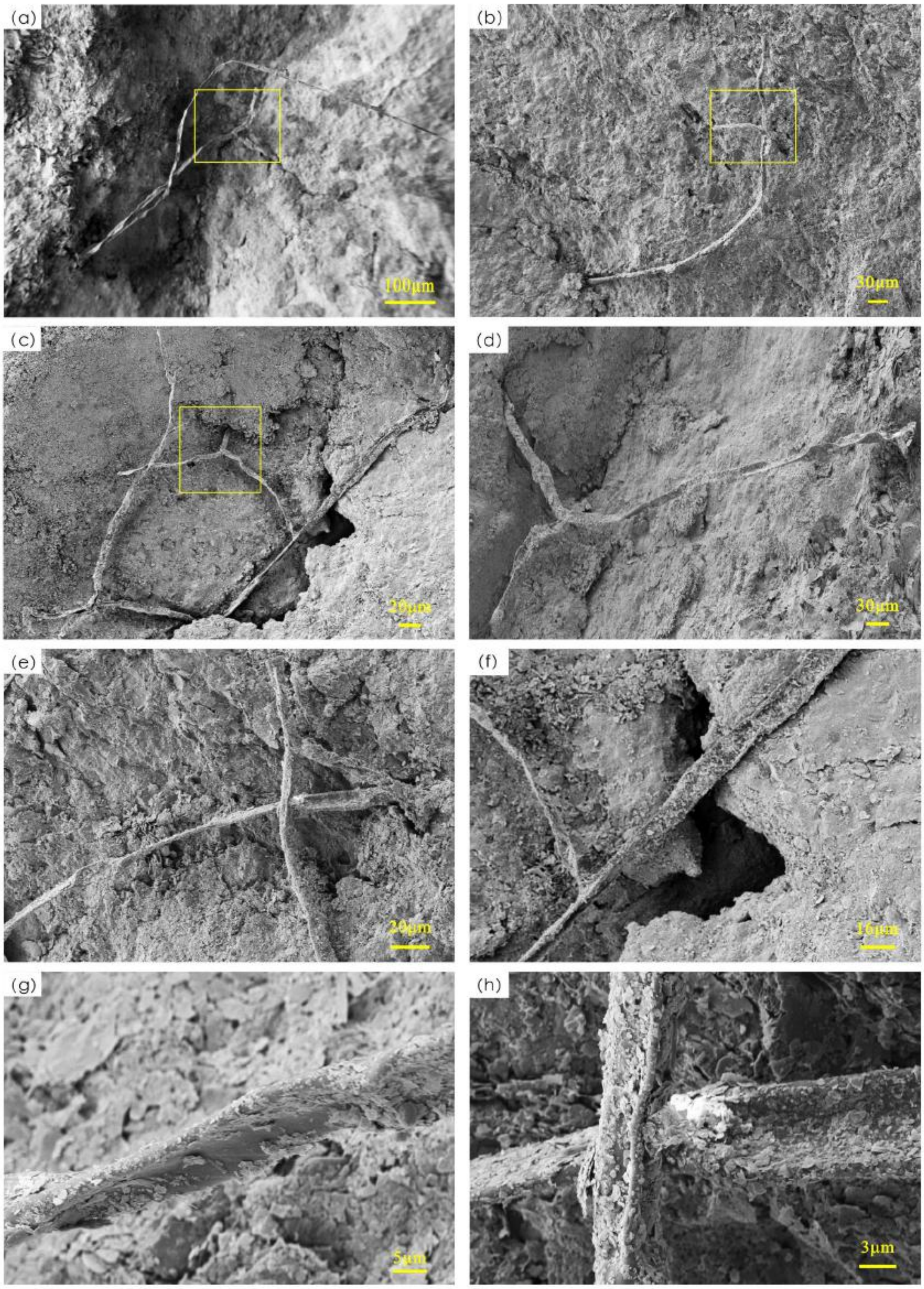


Fig. 5 SEM photos of the filamentous cyanobacterial fossils in a ferric stromatolite sample from the base of the terminal Paleoproterozoic Chuanlinggou Formation at Pangjiabu, in Xuanhua-Longguan area, Hebei province, China. (a): a squashed filament with a furcation (in the yellow box), approximately 1.2 mm in length and 13.3 μm in diameter. Filament No. PJBhfz9-a-1-003. (b): a squashed filament with a furcation (in the yellow box), approximately 300 μm in length and about 8.6 μm in diameter. Filament No. r-PJBhfz9-a-1040. (c): a squashed filament with a furcation, about 60-100 μm in length and 7.5 μm in diameter. Filament No. PJBhfz9-a-1-014. (d): a squashed filament, about 450 μm long and 18.8 μm wide. Filament No. r-PJBhfz9-

a-1-033. (e): a squashed filament, about 180 μm long and 5 μm wide. Filament No. r-PJBhfz9-a-1-052. (f): a squashed filament, about 60-100 μm long and 8.3 μm wide. Filament No. PJBhfz9-a-1-023. (g-h): r-PJBhfz9-a-1-035 and r-PJBhfz9-a-1-053, respectively, and their thickness is about 0.5-3.3 μm.

Table 1 Measurements of the filamentous fossils

Series number	Series number of the filamentous fossils	Length (μm)	Diameter (μm)	Furcating or not
1	PJBhfz 9-a-1-003	600-900	13.3	Pseudobranched
2	PJBhfz 9-a-1-005a	42.4-229.7	7.1	Pseudobranched
3	PJBhfz 9-a-1-005b	122.3	5.7	/
4	PJBhfz 9-a-1-005c	186.7	6.7	/
5	r-PJBhfz 9-a-1-013	100-923	7.5	Pseudobranched
6	r-PJBhfz 9-a-1-033	131.3-562.5	18.8	overlay
7	r-PJBhfz 9-a-1-037	471.4	12.9	/
8	r-PJBhfz 9-a-1-040	64.3-450	8.6	Pseudobranched
9	r-PJBhfz 9-a-1-049	642.9	10	/
10	r-PJBhfz 9-a-1-051a	495.9-527	3.9-9.3	overlay
11	r-PJBhfz 9-a-1-051b	411.5	6.6	overlay
12	r-PJBhfz 9-a-1-051c	65.2-463.6	5.9-11.3	/
13	r-PJBhfz 9-a-1-054	1000	26.7μm	Pseudobranched

ments (Fig. 5: (d), (e), (f)), indicating they are abundant. Some filaments have pseudobranching (Fig. 5: (a), (b), (c)). No filament shows obvious change in diameter within the exposed parts. All filaments expose a part of them, and their full lengths cannot be obtained. The lengths of the exposed parts of the filaments are generally hundreds of microns, with some up to 1 to 2 mm. Their diameters range from 3.9 to 26.7 μm (Table 1). The sizes of the filaments were measured and listed in Table 1.

4.3 Identification of the filamentous fossils

The fossils are all filamentous and less than 50 microns in diameter, belonging to microorganisms. Microorganism refers to microscopic organisms, including bacteria, cyanobacteria, viruses, archaea, fungi, microscopic algae, etc.

Bacteria are unicellular microorganisms, and can be morphologically divided into coccolithids, bacillithids and spiraithids. The coccolithids are spherical, and are generally about 0.5-2 μm in diameter. The bacillithids are rod-like, about 1.5-10 μm long and 0.5-1 μm in diameter. The spiraithids are long and spiral. Individual bacteria are very small. Rod-like bacteria are generally less than 1 μm in diameter and less than 4 μm in length. The lengths of the filamentous fossils found in this study range from hundreds of microns to several millimeters, and their diameters are generally 7.5-18.8 μm. Both of the length and diameter of the filaments are much greater than those of bacteria. So, they cannot be bacteria.

Archaea are mostly spherical, rod-shaped, spiral-shaped and irregular in shape, with diameters generally less than 1 μm. The fossils in this study are filamentous, are much greater than archaea, and their shapes are different from archaea. Thus, they are not archaea.

Fungi are generally branched and have complex branches. The filaments fossils obtained in this study are different from fungi in shape.

Cyanobacteria, also known as blue algae, are unicellular prokaryotes that can perform oxygen-producing photosynthesis. They generally have a sheath or are in EPS (extracellular polymeric substance), and can be morphologically divided into four types: spherical, irregular, unbranched filamentous, and branched filaments. The diameter of filamentous cyanobacteria is generally 0.5-50 μm.

The filamentous fossils in the iron formation are similar to cyanobacteria in several aspects: (1) Their diameters range from 3.9 to 26.7 μm, falling in the diameter scope of cyanobacteria, (2) They are fossils of sheaths (EPS), and sheaths are common in cyanobacteria but not in green algae, (3) The angle between the branch and the stem in the branching fossils is obtuse, close to 90°, similar to the case in extant cyanobacteria, and (4) Their diameters are stable for each filament, which is similar to the case in cyanobacteria but not to the case in extant filamentous green algae whose diameters obviously increase from base to end.

Filamentous species in green algae (Chlorophyta) belong to Ulotrichales, Chaetophorales, Oedogoniales and Cladophorales. Unbranched filamentous chlorophytes are morphologically similar to unbranched filamentous cyanobacteria. Their differences are: (1) the filamentous green algae of chlorophyta commonly do not have a sheath (EPS), while the filamentous cyanobacteria generally have a sheath (EPS); (2) Green algae are mainly in freshwater, with some in sea waters, while cyanobacteria are rich in both marine water and freshwater; (3) Most green algae have a holdfast, while cyanobacteria do not have; (4) Most filamentous green algae change greatly in their diameters, while most cyanobacteria have no obvious changes in filament diameter; (5) The angle between the branch and the stem in green algae is generally acute, but that in cyanobacteria is generally obtuse, often close to 90°. The five differences indicate that the filamentous fossils are more likely to belong to cyanobacteria than to green algae.

The sheaths of cyanobacteria are more difficult to degrade than cells and cell walls of cyanobacteria. We observed in our experiment that many sheaths were left after the cyanobacterial cells degraded. Therefore, we speculate that the filamentous fossils formed from the sheaths left by degradations of cyanobacterial filaments in two steps (Fig. 6).

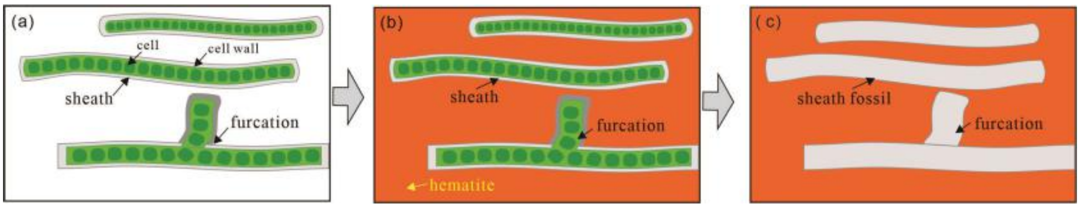


Fig. 6 Diagram illustrating the formation of the filamentous fossils. (a): Living cyanobacterial filaments in water. (b): Cyanobacterial filaments became embedded in hematite. (c): The cells of the filaments disappeared and their sheaths were preserved in hematite.

5. Significance of the cyanobacteria fossils

Microbial fossils are significant in determining the formation mechanisms and environments of ancient microbial rocks. Occurrence of abundant cyanobacteria sheath fossils in the Xuanlong-type iron formation confirms the existence of abundant cyanobacteria in the water from which the Xuanlong-type iron formation formed.

Cyanobacteria, as a group of photosynthetic autotrophs, can live in anoxic environment, and are generally distributed in marine photic zone. Their photosynthesis can produce oxygen. The absence of negative Ce anomaly, positive Fe isotope and relative enrichment of U, V and Mo in the Xuanlong-type iron formation indicate that the water of the iron formation might be hypoxic or anoxic, or a stratified ocean (Li et al., 2012). The presence of cyanobacteria fossils in the Xuanlong-type iron formation indicates that the formation water of the iron formation was within the ancient photic zone, and the cyanobacteria might have provided oxygen for the formation of the iron formation.

6. Conclusion

The stratiform iron formation at the base of the terminal Paleoproterozoic Chuanlinggou Formation in Hebei Province, China, i.e., the Xuanlong-type iron formation, consists of ferric stromatolites and ferric oolites. The ferric stromatolites are usually about 0.5-1 cm wide, and the ferric ooids are generally 0.1-0.3 cm in diameter. As found in this study, sheath fossils of filamentous cyanobacteria are locally abundant in the ferric stromatolites. The filamentous fossils are generally hundreds of microns to several millimeters long and range from 3.9 to 26.7 μm wide. Some of the filaments are branched. According to their morphology and size, the unbranched filamentous fossils are assigned into Osillatoriales of Cyanobacteria. The existence of the filamentous cyanobacterial sheath fossils indicates that the formation environment of the Xuanlong-type iron formation was within the ancient marine photic zone, and cyanobacteria may have provided oxygen for the oxygenation of the divalent iron ions in the seawater.

内容亮点

（1）用扫描电镜对采自中国河北省古元古代末（1650Ma）的铁质叠层石的自然断面进行观察，发现了直径3.9~26.7 μm、像尼龙丝一样的丝状化石；（2）化石的大小和形状与蓝细菌最为相似，所以认为它们是蓝细菌的胶鞘化石。

详细摘要

中国河北宣化-龙关一带的古元古代末长城系串岭沟组底部普遍发育数米厚的层状铁矿，被称做宣龙式铁矿，主要由赤铁矿组成的鲕粒和小型叠层石组成。铁质叠层石单个柱体的直径通常在0.5~1cm左右，铁质鲕粒的直径一般在0.1cm左右，较大的能达到0.2~0.3cm。在研究的三岔口（40°48′05.01″N，115°40′27.76″E）、李什沟村（40°74′9.57″N，115°63′93.02″E）和庞家堡（40°48′05.01″N，115°40′27.76″E）3个剖面上，串岭沟组由最底部的铁矿层和其上的黑色页岩、泥岩、粉砂岩组成。铁矿层在3个剖面上的厚度分别为2.5m、5m、7m，黑色岩系在3个剖面上的厚度分别为35m、75m、100m。铁矿层的下伏地层是常州沟组顶部的石英砂岩。串岭沟组的上覆地层是团山子组的白云岩。锆石测年获得的串岭沟组底界和顶界的年代分别是1650Ma和1640Ma，因此，宣龙式铁矿的时代大约是1650Ma。

本文作者对宣化一带3个剖面串岭沟组底部的铁矿层进行了野外观察、测量、取样。对其中5块样品的新鲜人工断面进行扫描电镜观察，在其中一块样品上发现了丰富的丝状化石。丝体化

石只在自然断面中出露了一部分；所见部分长度为数百微米至1~2毫米，直径在3.9~26.7 μm。各个丝体所见部分的直径稳定，无明显变化。部分丝体有分叉现象。分叉的丝体之间的夹角接近90°。将丝状化石的大小和形状与细菌、真菌、蓝细菌等主要微生物类群对比，发现它们与蓝细菌最为相似，因此认为它们是蓝细菌胶鞘的化石。现代蓝细菌生活在透光带。这些蓝细菌胶鞘化石的发现，说明宣龙式铁矿形成于浅水透光带，蓝细菌可能为铁矿的形成提供了氧气。

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