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Phosphorus Sources and Enrichment Mechanism of the Cambrian Xinji Formation, Baofeng, Western Henan

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Abstract

Large phosphorus deposition periods in geological history are often accompanied by major events such as global climate change, changes in atmospheric oxygen levels, and biogenic eruptions. The Xinji Formation of the Cambrian at the North China Craton southern margin is an important phosphorus-bearing formation in the north. In this paper, a typical phosphorus-bearing section of the Xinji Formation middle part in Baofeng, west Henan, was selected for study. The phosphorus-bearing section of the Xinji Formation was investigated using field section measurements, optical microscopy. The results show that the phosphorus nodules of the Xinji Formation are black in colour, vary in morphology and size, and their grain size varies from 0.2 cm to 2 cm, and they are distributed in fine sandstones along the facies; the main mineral of phosphorus nodules is colloidal phosphorite and some of them form oolitic colloidal phosphorite in the form of phosphatic shells of quartz grains. Deep-sea phosphorus-rich waters and small crustal organisms provide the material basis for phosphorus nodules in the Xinji Formation in the study areawhere colloidal phosphorites were deposited with quartz and other minerals in the early stages of deposition, and oolitic colloidal phosphorites were formed in the pores of the rocks in the early stages of diagenesis. The study of the phosphorus-bearing stratigraphy of the Xinji Formation can provide richer evidence for the recovery of the early Cambrian paleoenvironment of the North China Craton southern margin.

Keywords

North China Craton; Xinji Formation; Phosphorus-bearing horizons; Phosphorus sources; Enrichment mechanisms.

1. INTRODUCTION

Phosphorus is a major element in Earth's history and one of the important elements governing the ecology and nutrient component cycling on the Earth's surface[1,2]. Three large phosphorus-forming periods in geological history occurred during the Ediacaran-Cambrian, Permian, and Late Cretaceous to Early Tertiary periods[3]. Corresponding to these three phosphorogenic periods, there were global climate changes[4], changes in atmospheric oxygen levels, and biogenic outbreaks[5], among other significant events. This shows that the formation of phosphorus-bearing sediments as well as phosphate deposits is inevitably linked to environmental change, and the study of phosphorus in sediments is important for reconstructing long-term feedback mechanisms between climate, environment and ecology, as well as global climate change. Ninety-five percent of global phosphate resources belong to sedimentary rocks[2], with the tidepool zone being the most suitable environment for phosphate deposition[6,7]. The study of marine phosphate massifs began in the mid-19th century, and there has been more controversy about the source and formation mechanism of phosphorus-rich sediments, summarized as follows: phosphate originated from① terrigenous

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material input: weathering of phosphorus-bearing parent rocks[1,8]; ② Hydrothermal sources[8,9]; ③ Biogenic or marine authigenic sources[10-12], where nutrient-rich waters at depths of 100-300 m brought about by upwelling are considered to be the main source of phosphorus for phosphorites[10]. Phosphate formed by① Biological: Chen Qiying (2000) classified biological phosphate formation into direct and indirect formation[13,14]. ② Chemical: Phosphate adsorption by mica-illite, iron-bearing colloids etc[10]. ③ Magmatism[15], deposition of phosphate followed by physical mechanical re-enrichment to form industrial phosphate deposits[16]. Currently, biogenesis is becoming an increasing focus of research.

The phosphorus-rich sediments in China were mainly formed during the Ediacaran-Cambrian period, and their scale is the third among the three phosphorus-forming events, mainly distributed in the south-eastern margin of the South China Craton[1]. Previous studies have developed a multi-factor, multi-stage enrichment model for phosphate massif deposits in terms of their structural configuration, mineral composition, geochemical characteristics and depositional environment[17-19], suggesting that the phosphate massifs of the South China Craton are mainly biogenic phosphate massifs, which were deposited by mycorrhizal algae aggregating phosphorus at the beginning of deposition, deposited in the subtidal zone, and enriched secondarily into industrial deposits by surface weathering and leaching[20,21]. During the same period, a set of phosphorus-bearing strata, the Xinji Formation and its equivalent, was also deposited on the southern margin of the North China Craton. The stability of the substrate of the North China Craton and the warm, shallow marine environment suitable for biological growth are the basic reasons for the formation of the more stable phosphate mineralisation in the Xinji Formation[22], which mainly develops sandy (gravelly) phosphate blocks[18]. The Xinji Formation is an important phosphorus-bearing formation in northern China. Previously, studies on the petrographic palaeogeography, paleontological remains and microbial genesis structures of the Xinji Formation were carried out, and trilobites such as Hsuaspis and Bergeroniellus, horizontal tubular relic fossils[23,24], as well as microbial mat growth structures and microbial mat destruction structures[25]were found in this formation. The microbial mat growth tectonics and microbial mat destruction tectonics, but phosphorus-bearing horizons were rarely addressed until Pan et al. (2020) found a large assemblage of phosphorus-bearing brachiopod genera in the Xinji Formation in the Shuiyu section of Rui Cheng County, Shanxi Province[26]. In this paper, we investigate the typical phosphorus-bearing stratigraphy of the Xinji Formation of the Cambrian Series II in the Baofeng, west Henan Province, of the North China Craton southern margin, using field section measurements, optical microscopy, scanning electron microscopy with energy spectroscopy and other methods to investigate its sedimentology and mineralogy. We discuss the phosphorus source and enrichment mechanism of the Xinji Formation, analyse and explore its sedimentary characteristics and depositional environment. Constraints for restoration of the southern margin of the North China Craton Early Cambrian paleoenvironment.

2. REGIONAL GEOLOGICAL OVERVIEW

The North China Cambrian includes two major rock systems, namely the Xinji Formation to the Xuzhuang Formation , which is characterized by the interactive deposition of clastic rocks and carbonates, representing the development stage of the carbonate Cratonau, and the Zhangxia Formation to the Upper Cambrian, representing the mature stage of the Cratonau[27]. The Cambrian Canglangpu Stage marine intrusion reached the southern and southwestern margins of the North China Craton and deposited a set of strata: in the area from Luonan to Shangxian in the Northern Qinling Mountains, it is called the Sanchuan Formation; in the area from western Henan and Zhongjiao Mountains to northern Weibei, it can be called the Xinji

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Formation; and in the middle part of the Helan Mountains, it is called the Suyukou Formation[28]. This set of formations is relatively thick, generally around 100 m to 150 m[29], and often overlies parallel unconformities on top of the Aurignacian moraines (e.g., Luohuan Formation, Sandao Crash Formation, Zhengmuguan Formation, etc.)[3,29], with a phosphorus-bearing and often conglomerate-bearing sandstone or sandstone at the base and a dolomitic tuff interbedded with tuff at the top, forming a sedimentary gyre[25]. Protolenide-like trilobites, commonly Hsuaspis, Bergeroniellus etc, in addition to some primitive brachiopods[30]. The Lower Cambrian Canglangpu Order is the main phosphorus-bearing formation in northern China, distributed at the southern edge of the North China Massif, and phosphorus-bearing formations include the Xinji Formation and the equivalent Houjiashan Formation, and the Suyukou Formation[31]. The phosphorus-bearing facies consists mainly of terrigenous clastic and carbonate rocks, and is a terrane type terrigenous-carbonate deposit[3]. The Xinji Formation in Henan Province is distributed along the north-westerly stripes of the Qinling and Dabie Mountain northern margin depressional belt[25,31]. It is thickest in Yangsizhuang, Ye County, up to 334 m, and thins significantly from south to north, where it is bounded by quartz sandstones that disappear and thick layers of brecciated dolomitic tuffs that occur with the Zhusha Cave Group[31]. The study area is located in the village of Tsubshuping, Baofeng County, Pingdingshan City, Henan Province, in the shallow - coastal sandstone siltstone phase. The measured section belongs to the middle section of the Xinji Formation, which is a clastic sediment dominated by fine and siltstone.

3. STRATIGRAPHIC FEATURES OF THE PROFILE

The measured section is located at the southern margin of the North China Craton, with a point of N33°57′49″ and E112°46′00″. Due to vegetation cover, the top and bottom of this group are not visible. The overlying strata are greyish white dolomitic tuff , integrated contact with the underlying Dongpo Formation lime green mudstone in parallel unintegrated contact. 16 samples were collected. The total thickness of the section is 2.733 m. Details of the measured section are described below.

The lithology of this section is mainly fine-grained sandstone, siltstone, siltstone and siltstone mudstone. The sediments become progressively finer grained from the bottom up, with positive grain order lamination. The lithology tends to be 115°, with a dip of 8°, and is nearly horizontally laminated. The sedimentary structure of the Xinji Formation in the study area is simple, with mainly blocky laminations and clear parallel laminations at the top. Phosphorus nodules are common in the Xinji Formation, especially in the grey-green fine sandstone. The phosphorus nodules are black in colour, smooth on the surface, clearly demarcated from the fine sandstone, varying in size and shape, often rod-shaped, ellipsoidal, spherical and flattened. Microscopic observations show that the quartz grains in the sandstone in the lower part of the section are mostly smaller, contact cemented, with a grain-supported structure, less cemented, well sorted, sub-angular-sub-rounded, with the presence of sea chlorite, a small amount of zircon, and iron fragments. From the bottom to the top of this section, the quartz grains become progressively finer, the inter-grain cementation increases, and the content of sea chlorite, zircon and iron fragments decreases, with occasional minor metamorphic sericite. The two lamellae at the top of the section show clear microscopically interspersed light and dark bands at irregular intervals, with quartz dominating the light band and mudstone minerals the dark band.

4. CHARACTERISTICS OF PHOSPHATIC SEDIMENTS

Optical microscopic observation of the phosphorus-bearing nodule sandstone flakes shows that the main manifestation of the phosphorus nodules is colloidal phosphorite, with the

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phosphatic colloidal structure being the main structure of the nodules. The enclosing mineral grains are mainly quartz, with a small amount of sea chlorite and iron fragments interspersed.

Colloidal apatite is not easily distinguished as a single particle of apatite, mainly in the form of cryptocrystalline aggregates distributed among quartz grains, commonly round and oval in shape, partly irregular, with clear mineral margins. Grain sizes are in the 5-10 mm range. The aggregates are mainly colloidal amorphous forms, with a small amount of microcrystalline apatite visible, with no internal structure, often wrapped around better sorted quartz grains. Colloidal apatite is brownish in single polarised light, colloidal apatite itself is colourless and transparent, turbid when containing traces of mud and organic iron [32], extinct in orthogonal light, with homogeneous anisotropy. In contrast to the phosphate microstructure of phosphate microorganisms from the Steep Hill Tuo Formation published by Yang (2019) [33] and others, the phosphorylated animal embryos from this group have closely arranged multicellular structures, indicating that microorganisms took up phosphate supplemental nutrients directly from the aqueous medium and were preserved by phosphorylation after death.

The oolitic structure of the colloidal phosphorites in this section has a special structure: oolitic and torrid. These bivalve thin-skinned ooids were generally formed in a low-energy environment [34] and show cross extinction in orthogonal light, probably as a pseudo-cross wave extinction caused by the coaxial arrangement of apatite. The phosphatic shell may have been precipitated from phosphorus in pore water in the form of fibrous bright crystals [17]. Tortoise cracks are formed when colloidal phosphorite underwent dehydration during diagenesis and are only seen in dense massive ores [35]. The irregular shape of the cracked geophosphate ore and the dense cracking in some of the geophosphate ores in this section indicate that the phosphatic deposits underwent diagenetic processes. The difference between the tortoise fissures and the phosphatic faunal embryos is that the latter have a distinct shell of cells with neat margins, and the cells of the phosphatic faunal embryos are significantly smaller. The microscopic morphology of phosphorus in the phosphorus-bearing strata of this section of the profile is shown in Table 1.

Table 1. Microscopic morphology of phosphorus of Xinji Formation

Sample number	Profile location	Microscopic form and structure of phosphorus
XJ-08	Light purplish-red chalky sandstone	Colloidal phosphate ore, oolitic structure, cracked
XJ-06	Grey-green fine-grained sandstone	Colloidal phosphorite, phosphatized animal embryos, tortoiseshell, oolitic structures
XJ-05	Upper section of purplish red fine-grained sandstone	Colloidal phosphate ore, oolitic structure
XJ-03	Lower to middle section of purplish red fine-grained sandstone	Colloidal phosphorite, cracked
XJ-02	Lower section of purplish red fine-grained sandstone	Colloidal phosphate ore

Scanning electron microscopy of the fine-grained sandstones of the Xinji Formation containing phosphate nodules shows that the phosphate nodules consist of a large number of amorphous apatites, which are produced as ovoid and spherical grains of almost identical size and shape, with a grain size of 1-2 μm , undirected and densely arranged, without pores, irregularly stacked, distributed between quartz grains, or occurring as massive aggregates, widely distributed, and are the main crystal form of colloidal phosphate ore. This feature is

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consistent with the phosphatic characteristics of primary deposits in seawater proposed. Other microfine grained apatites form microlaminations around quartz grains, which exhibit ring edges of quartz grains. The different crystallographic patterns exhibited by the apatites indicate different depositional environments during the period of phosphatic deposition. In all the observed phenomena, no fissures and pores resulting from accountancy are visible. It can be concluded that the phosphorus nodules did not undergo significant accountancy modification, but are characterized by primary formation.

5. DISCUSSION

5.1. Phosphorus Sources

Late Aurignacian-Early Cambrian phosphate formation is closely related to sea erosion, and the formation of phosphorus-bearing rock systems during the Early Cambrian phosphate formation was also related to global sea-level changes[36], and Liang et al. (2019) analysed the geochemical characteristics of phosphorus-bearing sandstones of the Ordos Xinji Formation, indicating that upwelling and sea erosion carrying phosphorus and organic matter may have been the main source of phosphorus-bearing sediments[12]. A comprehensive analysis of paleontological and geotectonic evidence suggests that the Middle Cambrian North China Craton may have been within the Paleopacific Ocean and located between the Lauren and East Gondwana continents, and paleomagnetic evidence suggests that the Middle Cambrian North China Craton was located near 20°N[37]. The melting of Neogene glaciers caused a continuous rise in global sea level during the Cambrian[38], and seawater invaded from the southern and southwestern margins of the North China Craton[39], and as the extent of the sea invasion expanded, oceanic circulation was enhanced by the action of the low-latitude westward currents[38], triggering Upwelling brings deep-sea phosphorus-rich water to shallow marine areas, providing a physical source for the formation of phosphorus nodules.

5.2. Enrichment Mechanisms

From the morphological features, phosphorus-rich deposits from the North China Craton southern margin are independently bedded as phosphate nodules, sandwiched between black lithologies or dolomites; phosphorus nodules of the Xinji Formation in the study area are deposited in fine sandstone lithologies with smooth surfaces, and no traces of accounting are found in the lithologies, indicating differences in depositional environments and material sources between the two areas. Microscopically, the phosphorus nodules appear mostly as colluvial-cryptocrystalline aggregates, representing primary deposition[34]. The absence of biological remains in the phosphate nodules in the study section and the presence of a few phosphate animal embryos indicate that microorganisms absorbed phosphorus from seawater for life and that biogenesis was indirectly involved in the formation of phosphate nodules, and that primary deposition, chemically, was still the main role in the formation of phosphate nodules. In all the observed phenomena, no fissures and pores resulting from accounting were seen, further verifying that the phosphates of the Xinji Formation came from seawater and were deposited under chemical action. In terms of crystallinity, the colloidal aggregates of colloidal phosphate are homogeneous and anodic in orthogonal light, while those with an oolitic structure show cross extinction; the widely distributed colloidal phosphate is more visually evident in SEM as irregular aggregates of 1-2 μm non-crystalline ovoid grains, while those in shell form show an ordered arrangement of microfine grained apatite. The degree of mineral crystallisation is controlled by temperature, pressure and solubility, suggesting that the phosphates in this group formed in different environments and at different times. The phosphate nodules were formed in the early diagenetic stage, and the quartz grains encapsulated within the glufosite were either aggregated at the edges of the glufosite or uniformly dispersed throughout the glufosite, suggesting that the glufosite and quartz were

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deposited simultaneously and that the phosphate began to be deposited at the early stage of sedimentation; the glufosite shell crust may have been precipitated from phosphorus in the pore water at the early stage of diagenesis[17]. During later diagenesis, the early-deposited colloidal phosphorite underwent dehydration to produce torrid fractures on the surface.

The macroscopic features, microscopic morphology and mineral assemblages of phosphorus nodules lead to the understanding of the genesis of phosphorus nodules: due to the geochemical cycling of phosphorus, deep seawater contains a large amount of dissolved phosphorus, providing the material basis for the formation of later phosphorus-rich deposits. Seawater invaded northwards from the southern and southwestern margins of the North China Craton in the early Cambrian, first reaching the depressional zone in the northern part of the Qinling-Dabie orogenic belt, and gradually invading the central part of the North China Craton. The upwelling of seawater brought phosphate from deep water to the surface of the water body, part of which was absorbed and fixed by microorganisms living at the surface and released through life activities, increasing the concentration of phosphate in the surrounding water body. At the same time, because of environmental changes such as reduced pressure, increased temperature, escape of CO_2 and increased pH of the water column, phosphate in seawater begins to be deposited due to chemical and bio-chemical interactions, and is later adsorbed by impurities such as organic matter to form various shapes, with the overall dark brown colour of the colloidal phosphate ore. In the early stages of diagenesis, as microorganisms died and were deposited in the sediment, phosphorus was released again by the decomposition of organic matter, increasing the concentration of phosphorus in the pore water of the sediment, and as the overlying rock layer thickened and the pressure increased, dissolved phosphate precipitated and wrapped around the periphery of the quartz particles.

6. CONCLUSION

Deep-sea phosphorus-rich waters and small crustal organisms provide the material basis for phosphorus nodules in the Xinji Formation in the study area. The macroscopic characteristics, microscopic morphology and mineral assemblage of phosphorus nodules lead to the understanding of the genesis of phosphorus nodules: at the early stage of deposition, upwelling brought phosphorus-rich water from the deep sea, and due to environmental changes and indirect enrichment by microorganisms, phosphate began to be deposited and was adsorbed by organic matter and other impurities to form dark brown colloidal phosphate ores of various forms. In the early stages of diagenesis, dead microorganisms were decomposed by organic matter to release phosphate, making the phosphate concentration in the sediment pore water larger, and as the overlying rock layers thickened and became more pressurised, phosphate precipitated and was deposited in the crevices of the quartz grains, forming a phosphatic envelope of quartz grains.

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