Nanoparticles in the Earth surface systems and their effects on the environment and resource

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ABSTRACT

Nanoparticles (NPs) in the Earth surface systems have been a global concern due to their abnormal effects and their scientific significance and application value in environmental remediation and resource exploration. By means of comparative and comprehensive studies: (1) A description of the development of techniques for detecting NP characteristics and comparisons between those techniques are summarized; (2) the characters of typical NPs, including minerals at the nanoscale, nano organic matter, natural metal NPs, nanoinclusions, and NPs in soil and water are concluded; (3) the physical, chemical, microbial, and multigeological origins of NPs are revealed; and (4) the abnormal properties of NPs including the size, migration, aggregation, and adsorption effects and oxidation–reduction reactivity, are elaborated upon.

The formation, migration, deposition, and accumulation of NPs in the Earth surface system are closely related to the environment evolution and resource enrichment, and therefore NPs have environment and resource effects. On this basis, the uses of NPs in environmental remediation and resource exploration are clarified: (1) uses of NPs and modified NPs and combinations of NPs and other remediation technologies are analyzed, and the main factors influencing the remediation effect, such as the soil characteristics, hazardous element species, and NPs themselves, are examined; (2) NPs originating from plutonic fluid and weathering can indicate the mineralizing mechanism and instruct the identification of concealed deposits. For gas and oil exploration, organic NPs can reflect hydrocarbon-generating processes and determine gas and oil occurrence states. Organic NPs are also widely involved in the mineralization process of metal elements and thus can indicate the coexistence characters of organic matter and metal.

1. Introduction

The Earth surface is a dynamic union of its solid crust, atmosphere, hydrosphere, and biosphere, all acting in concert to produce a constantly renewing and changing symphony of form (Pieri and Dziewonski, 2007). Nanoparticles (NPs) are nanosized (usually < 100 nm) particles for which many physical and chemical properties (such as adsorption, dissolution, aggregation and chemical reactivity) differ from those of macroscopic objects (Hochella, 2002; Hochella, 2008; Ju et al., 2017). In the Earth surface systems, which provide essential resources for the sustainable development of human societies, various forms of NPs are found (Yang et al., 2018). For example, NPs are increasingly found in geologic environments including mineralization (Palenik et al., 2004), oxidation and reduction (Hough et al., 2008; Griffin et al., 2018), fault activities (Wilson et al., 2005; Hu and Cao, 2019), microbial actions (Lengke and Southam, 2006), weathering (Wada, 1987; Reith et al., 2012; Xu et al., 2017), volcanic eruptions (Lähde et al., 2013), and submarine jets (Wang et al., 2013). NPs in geological environments can be excellent indicators of the mechanisms underlying geological processes and are used for the exploration of resources (Li and Pan, 2015), considering that NPs may carry many chemical or physical or biological information about the deeply buried strata or ores. Polluting NPs originating from geological or artificial processes can be found in soils and waters (Kretzschmar and Schäfer, 2005). Due to the effects of NPs on

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resources and environments, attention to NPs is growing for their roles in exploring concealed resources (Hochella and Madden, 2005; Theng and Yuan, 2008; Hu and Cao, 2019) and in disposing contaminants (Fu et al., 2012; Ju et al., 2017).

Nanotechnology developments have provided new ideas and insights for humans to solve critical problems in the environmental, biological, and energy fields (Wirth, 2004; Bhushan, 2005; Porter and Youtie, 2009). At the beginning of the 1980s, geologists introduced nanotechnology-based geological tools and defined the basic concept of nanogeoscience (Binnig et al., 1985). Continuous progresses have been achieved over the subsequent 40 years, and several branches of nanogeoscience have been formed, such as nanomineralogy (Gao et al., 2014, 2016; Bergström et al., 2015; De Yoreo et al., 2015; Sturm and Cölfen, 2016; Huang et al., 2017), nanopetrology (Wu et al., 2002; Qiu et al., 2003; Ju et al., 2005), nanostructural geology (Ju et al., 2005), nanogeochemistry (Wang et al., 2012, 2016a, 2016b; Zhang et al., 2018), nanomineral deposits (Wang et al., 2007; Cao et al., 2009, 2010b), nanomarine geology (Wang et al., 2013; Guan et al., 2017, 2019; Wu et al., 2016; Li et al., 2020a), nanoenergy geology (Ju et al., 2005; Loucks et al., 2009; Curtis et al., 2012a, 2012b; Zou et al., 2017), nanoearthquake geology (Wilson et al., 2005; Viti and Hirose, 2010), and nanoenvironmental geology (Shao et al., 2007; Buseck and Adachi, 2008; Theng and Yuan, 2008; Wang et al., 2011). NPs are the key to deepen the scientific understanding of these branches of nanogeoscience, particularly in revealing geological and environmental evolution processes and mechanisms in different layers of Earth (Ju et al., 2017), by studying their origin, migration, deposition and evolution. Natural NPs are often overlooked compared with engineered NPs (Griffin et al. 2018), and the formers require more attention (Liu et al., 2020b). Variations in the shape and composition of natural NPs are generally more complex than those of engineered NPs, considering that the latter is generally more uniform than is the former (Lima et al., 2021; Oliveira et al., 2021a; Ramírez et al., 2020; Silva et al., 2021a).

Based on quantum and surface effects, the most essential characteristic of NPs is that their special physical and geochemical properties, which differ from their macroscopic counterparts (Hochella, 2008), thus giving rise to the complexity of the formation, migration, aggregation, and evolution mechanisms of NPs (Ju et al., 2018). Geological processes that involve nucleation and crystal growth, precipitation, dissolution, phase transformation, surface reactivity, and mineral–microbe interactions are all related to nanoscale phenomena (Reich et al., 2011). The special atomic arrangement and surface properties of NPs make NPs have properties different from those of the macro particles (Banfield and Zhang, 2001). As crystal size changes from the micron to nanometer scale, the basic physical and chemical properties of solid materials (processing the same chemical composition and crystal structure) change by multiples or even orders of magnitude (Liu et al., 2018), thus creating difficulties and challenges for researching geological or environmental interactions and evolutionary processes between NPs and other media or the NPs themselves (Fu et al., 2018). Accordingly, although NPs generated from geological or environmental processes have been substantially detected, precisely observed, quantitatively or quantitatively analyzed, and comprehensively summarized by advanced technical methods, the overall understanding of NPs' characteristics in various geological and environmental conditions in the Earth surface systems and their influences on resource exploration and pollution remediation remain imperfect (Reich et al., 2006; Hochella, 2008; Theng and Yuan, 2008; Hassellöv and Kammer, 2008; Bargar et al., 2008; Reich et al., 2011; Ju et al., 2017; Hochella et al., 2019).

Unprecedented scientific discoveries can be achieved by exploring the special properties, formative conditions, action processes, migrations, and aggregation laws of NPs. Consequently, the newly developed interdisciplinary nanogeoscience shows great vitality and broad prospects, thus representing new revolutionary progresses at the microscale of geoscience (Hochella, 2002, 2008; Hochella et al., 2008; Wang, 2014c; Ju et al., 2017). The research techniques, special properties, and origins of NPs are systematically introduced. The current research status of typical NPs in the Earth surface systems are also elaborated. Combined with nanomineralization, the special migration and accumulation properties of NPs in geogas and ore-forming hydrothermal fluids while discussing the indicative significance of NPs in the exploration of concealed deposits have been introduced. In addition, the main NPs and technologies applied in soil and water hazardous element pollution modification and the main factors influencing the application effect are also presented. Ultimately, the applications of NPs in soil and water remediation and resource exploration are investigated and summarized.

2. Research methods of NPs

Important information such as the structure, morphology, mineral phase, and chemical composition of NPs can be provided by advanced techniques which are summarized as follows. The development of the first scanning tunneling microscope (STM) in 1982 made nanoscale observations possible based on the scanning tunneling effect, accelerating the development of nanotechnology (Ju et al., 2016). Rapid advances in high-precision microanalytical techniques over the past 20 years have provided a range of incredible opportunities to understand the morphology, crystal structure (even at the atomic scale), and element distribution of natural NPs (Reich et al., 2011; Li and Pan, 2015). Using these advanced techniques, studies on NPs have shed light on the crystallization and growth processes of minerals, thus offering insights into the Earth's geological evolution processes (Hochella et al., 2008; Li et al., 2013; Ju et al., 2017). For example, using high-pressure freezing FIB-SEM technology, Schmid (2014) found that iron oxidizing bacteria could simultaneously form nano-sized iron minerals on the cell surface and cell membrane. The double-layer mineralization between the cell surface and cell membrane wraps iron oxidizing bacteria via iron mineralization, forming "mineral bacteria" inclusions, which is conducive to the preservation and final formation of microfossils that can be used to analyze the Earth's geological evolution processes.

Aided by continuous developments in nanoscience and nanotechnology, humans' understanding of natural (Li and Li, 2016; Ju et al., 2017; Hochella et al., 2019) and incidental (Akinyemi et al., 2019; Duarte et al., 2019; Ferrari et al., 2019; Gasparotto et al., 2019) and engineered NPs (Lanknoe et al., 2017; Praetorius et al., 2017; Hochella et al., 2019) is deepening. Optical microscopy (OM), which was initially applied to the study of geology in the 1820s (Howard and Dawn, 2014; Tang and Li, 2021), allowed humans to study the micro world. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM), which play important roles in researching morphological characteristics, crystal structures, mineral phase identification, chemical composition, atomic imaging, and micro magnetic structure from the micro- and nanoscale to the atomic level for natural NPs, were developed in the 1930s (Borries et al., 1938; Kauschi et al., 1939) and 1940s (Oatley, 1982), respectively. Since the 1960s, secondary ion mass spectrometry (SIMS) and nanosecond ion probe mass spectrometry (NanoSIMS) have allowed researchers to obtain element and isotopic information for natural NPs (Hoppe et al., 2013; Yang et al., 2015). Atomic force microscopy (AFM) and STM emerged in the 1980s, offering a high resolution of 0.01–0.1 nm (Binnig and Rohrer, 1985) and providing quantitative measurements of NP morphology at the atomic scale (Tang and Li, 2021), thus making

atomic-scale exploration of natural NPs a reality. As the use of atom probe tomography (APT), Frierdich et al. (2019) reacted nanoparticulate goethite and used APT to resolve the three-dimensional distribution of Fe isotopes in goethite, concluding that NP encapsulation and subsequent APT can capture hidden recrystallization mechanisms which are critical in predicting mineral reactivity in aqueous solutions.

The advantages of different analytical techniques for detecting the characteristics of NPs are summarized in Table 1, and the spatial resolutions and detection ranges for such techniques are displayed in Fig. 1.

Combinations of different techniques for researching natural NPs have flourished since the beginning of the 21st century, allowing for combined research on the morphology and composition of natural NPs (Yan et al., 2018; Zhou et al., 2021). For example, the use of a focused ion beam (FIB) gives control over the size of the prepared samples for NP analysis to ensure a uniform thickness suitable for a variety of microscopic and microspectral analyses (Oliveira et al., 2018; Sánchez-Peña et al., 2017), such as SEM, TEM, NanoSIMS, and synchrotron radiation scanning transmission X-ray microscopy (STXM). On this basis, FIB–SEM–TEM, FIB–SEM–atomic probe tomography (APT), and even FIB–TEM–STXM combinations have offered more comprehensive multiscale and multidimensional information about specific nanoparticles. For example, Pokrovski et al. (2019) applied high-energy resolution fluorescence–detected X-ray absorption spectroscopy (HERFD–XAS) with traditional analytical techniques, such as SEM, X-ray diffraction (XRD), electron probe microanalysis (EPMA), laser ablation inductively coupled plasma mass spectrometry (LA–ICP-MS), and inductively coupled plasma atomic emission spectrometry (ICP-AES), to clarify the nanogold redox and structural state during interactions between gold-bearing sulfur-rich hydrothermal fluids and arsenic-free pyrite at high temperatures (350–450 °C) and high pressures (400–700 bar). They found that dissolved Au in sulfide-sulfate solutions forms complexes with hydrogen sulfide. In particular, invisible gold in pyrite occurs as Au metal submicron- to nano-
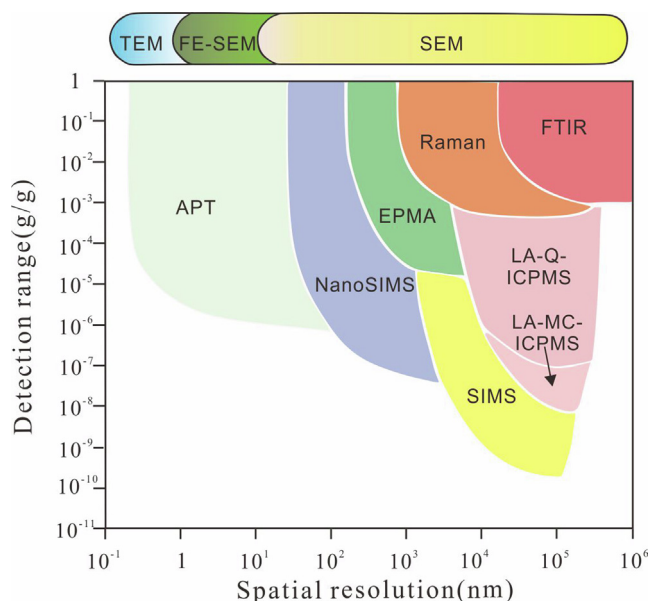

Fig. 1. Spatial resolutions and detection ranges of distinct analytical techniques (modified from Li and Li (2016)).

sized particles and chemically bound Au^+ in the form of (poly) sulfide clusters composed of S-Au-S linear units (Pokrovski et al., 2019).

The rapid development of techniques for researching NPs has greatly improved the understanding of natural NPs, and certain factors must be considered. Natural NP samples are usually highly heterogeneous and poorly crystallized, and techniques for observing NPs are generally performed under high voltages, which can readily cause radiation damage or amorphization of the samples. These two factors can change the intrinsic structure of samples and eventually lead to error. To reduce such negative effects, testers experienced with sample preparation, experimental operation, and parameter adjustment are required. Moreover, research into NPs cannot be divorced from atomic- and macroscale research. Nanoscale and larger- and smaller-scale technologies must be combined for multiscale analysis. Techniques for image observation (e.g., SEM, TEM, STM), component analysis (e.g., XRD, FTIR, LA-ICPMS, and ICP-AES) and, advanced sample operation (e.g., FIB and argon ion polishing) must be combined to comprehensively research NP morphology, components, structure, and origin (Ramos et al., 2015; Civeira et al., 2016; Dalmora et al., 2016).

3. Research status of typical NPs in the Earth surface systems

Bernhardt et al. (2010) classified NPs into naturally occurring NPs, secondary (or manmade) NPs, and engineered NPs according to their origins. Nanosheets, nanorods, and NPs have one, two, and three dimensions in the nanorange, respectively (Hochella, 2008). The research status of various typical NPs in the Earth surface systems, including nanominerals (Hochella, 2008; Hochella et al., 2008), nanoinclusions (Verdugo-Ihl et al., 2019), nano organic matter (Ju et al., 2017), natural metal NPs (Saunders et al., 2017; Han et al., 2020), and natural metal nanoparticles (Zou et al., 2016), was summarized and discussed to gain insights into typical NPs.

3.1. Minerals at the nanoscale

Minerals at the nanoscale can be classified as nanominerals and mineral NPs (Hochella et al., 2008). The former are minerals that

Table 1
Advantages of different techniques for detecting NP characteristics.

Techniques	Advantages
STM	NP crystal structure, unit cell size and shape, and crystal defects can be observed using STM, and formative and evolutionary information can be clarified.
TEM	NP morphology, mineral phase, crystal structure, and chemical composition can be comprehensively observed and analyzed.
SEM	NP surface morphology can be observed using SEM by integrating various imaging techniques to take advantage of intuitive imaging, high resolution, long depth of field, strong stereoscopic effects, and convenient sample preparation. SEM also allows for in situ and real-time observations.
AFM	Atomic resolution of the surface structure and morphology of NPs can be achieved using AFM.
EPMA	Elemental composition and distribution as well as mineral trace elements on NP aggregates surfaces can be analyzed.
NanoSIMS	In-situ analysis of isotope and trace elements, scanning imaging of NP elements and isotope distributions
LA-ICP-MS	Accurate analyses of the distribution of elements and isotopes of NP aggregates can be realized, and composition analyses of single-fluid inclusions and U-Pb dating of accessory minerals are also possible.
APT	Boundaries between different crystals and phases, as well as NP structures and their dislocations and defects can be observed using APT. The heterogeneous distribution of elements and isotopes of accessory minerals can also be analyzed on the nanoscale using this technique.

Abbreviations: scanning tunneling microscope (STM), transmission Electron Microscope (TEM), scanning electron microscope (SEM), atomic force microscope (AFM), electron probe microanalyzer (EPMA), nanosecond ion probe mass spectrometry (NanoSIMS), laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS), atom probe tomography (APT).

exist only in the nanosized range (<100 nm), such as certain clays as well as iron and manganese (oxyhydr) oxides, whereas the latter are minerals that can also exist at larger sizes and probably including most known minerals (Hochella et al., 2008). The differences between nanominerals and mineral NPs and NPs are introduced as follows: minerals at the nanoscale can be classified as nanominerals and mineral NPs (Hochella et al., 2008). The former are minerals that only exist in the nanosized range (<100 nm) while the latter are minerals that can exist on the nanoscale and at larger sizes. NPs not only include nanominerals and mineral NPs, but also include other NPs such as nanoinclusions and nano organic matter. Thus, "minerals at the nanoscale" contain nanominerals (can only exist at the nanoscale) and a part of mineral NPs with at least one dimension at the nano range. Minerals at the nanoscale are limited to one, two, or three dimensions and can produce nanorods (e.g., nanotubular halloysite); nanofilms or nanosheets (e.g., nanoclay minerals), or NPs (e.g., nanogold aggregations) (Hochella et al., 2008; Ju et al., 2017). Nanominerals' and mineral NPs' physical and chemical properties change with size and morphology (Ju et al., 2017). Nanominerals have diverse forms and widely exist in the Earth surface systems and even in celestial bodies (Fu et al., 2018). Under the action of biological or abiotic processes, the formation of minerals will go through the nanophase stage, which is usually temporary (Penn and Banfield, 1998). Fig. 2 displays examples of some minerals at the nanoscale in ore deposits (Fig. 2a), soils (Fig. 2b), water bodies (Fig. 2c), and meteorites (Fig. 2d).

3.2. Nano organic matter

Research into nano organic matter (kerogen) has developed rapidly with the global boom in unconventional gas and oil. The advanced experimental techniques introduced in Section 2 have been widely used to investigate the morphology, distribution, type, and hydrocarbon generation potential of nano organic matter in shale or tight reservoirs (Ju et al., 2017; Lu et al., 2017). In recent years, the unique macromolecular structure characteristics of organic matter have been studied because organic macromolecules can reflect the type, source, sedimentary environment, and metamorphic deformation of organic matter, thus providing an important basis for inversion of the entire geological process (Ju et al., 2017). However, the characteristics and models of hydrocarbon generation of nano organic matter may differ from conventional kerogen because of its special nanometer effect. Additionally, the combination of complex distributed dispersed or concentrated nanokerogen and inorganic nano- or macrominerals with heterogeneous wettability may impede the production of gas or oil (Hu et al., 2018b; Zhao et al., 2020). This impediment occurs because the mobility of gas or oil in those irregularly distributed organic or inorganic nano-structures are quite heterogeneous, leading to the discontinuous flow of gas or oil, thus requiring the study of the movable gas and oil reservoir space and flow capacity of different lithofacies influenced by nano organic matter and nanominerals.

3.3. Natural metal NPs

Natural metal NPs have been found in ores of metal deposits, such as gold (Su et al., 2008; Saunders et al., 2017; Hu et al., 2018a) and uraninite (Bonnetti et al., 2015; Schindler et al., 2017). They can also be found in polymetal deposits such as Cu–Ni (Mikhlin et al., 2017; Mavrogonatos et al., 2019). Moreover, by means of using TEM, NPs of crystals (e.g., native copper, gold, and alloys such as Cu–Fe and Cu–Fe–Mn and Cu–Au) were observed in gases, soils, and ores (Wang et al., 2016a, 2016b). In gold deposits, for example, a significant amount of gold occurs as
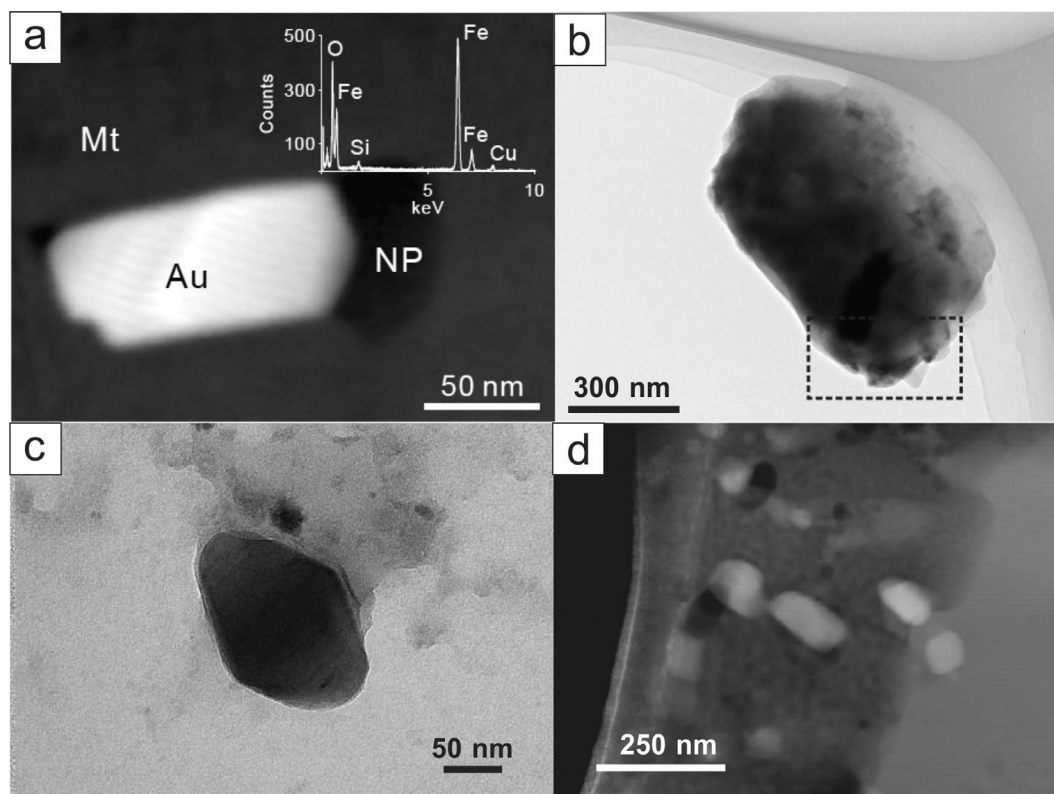

Fig. 2. (a) High angle annular dark field (HAADF) images showing gold NPs in low-Si magnetite from Zhou et al. (2021); (b) gold-bearing NPs in soil samples from Zhang et al. (2019); (c) TiO$_2$ NPs in water from Cheng et al. (2018); and (d) NPs in chondrite named DaG 978, which are displayed in a HAADF-STEM image from Li et al. (2017).

discrete nanograins disseminated in gangue silicate minerals (Saunders et al., 2017), as nanogold (Au^0) in pyrite or arsenian pyrite (Reich et al., 2005; Su et al., 2008; Hough et al., 2011), or as "invisible gold" in clays, iron oxides, and carbonates (Hough et al., 2011). In sandstone-type uranium deposits, significant amounts of uranium are adsorbed on clay mineral surfaces as U^{6+} ions or reduced, mainly as UO_2 nano- or micro-crystals disseminated in the clayey matrix (Bonnetti et al., 2015; Wang et al., 2016a, 2016b; Schindler et al., 2022). Natural polymetallic NPs are 50–700 nm in size and contain elements that are among the most abundant in the Earth's crust, such as O, Mg, K, Ca, Si, Al, Ti, Fe, and P (Liu et al., 2020a), along with precious metal elements, and have concealed ore body-related elemental associations, such as S–Cu, S–Zn, S–Pb, S–Mo, S–Fe–Zn, S–Fe–Cu, S–Fe–Cu–Mo, S–Fe–Zn–Pb, and Cr–Mn–Fe (Filimonova and Trubkin, 2008; Luo et al., 2015; Schindler et al., 2022). In recent years, natural metal NPs have been widely observed in geogas samples from different kinds of concealed deposits (Fig. 3) (Wang et al., 2007, 2012; Cao et al., 2009, 2010a, 2010b; Zhang et al., 2019; Han et al., 2020). The mechanism underlying the upward migration of these natural metal NPs from buried deposits through the cover to the surface remains controversial (Zhang et al., 2019). One hypothesis is that natural metal NPs or metal elements are carried onto the bubbles or microflows of geogas and migrate to the surface (Wang et al., 2007, 2017b). Natural metal NPs transported by geogas flows ascending from deeply concealed deposits potentially hold great ore formation information (Jiang et al., 2019). The category, size, shape, chemical composition, and structure of natural metal NPs have been investigated in previous works (Wei et al., 2013).

3.4. Nanoinclusions

Inclusions, for example, fluid inclusions, which are the most direct record of the geochemical and physical properties of ancient fluids trapped deep in the Earth's crust, provide essential information about the geological formation of hydrothermal ore deposits (Lin et al., 2020). Nanoinclusions that reflect ore formation information have been found while researching ore genesis and evolution. Verdugo-Ihl et al. (2019) examined HAADF STEM images of the distribution of nanoinclusions along trails in hematite and found that the finest inclusions (<20 nm) were distributed as a continuous array (Fig. 4). The components of nanoinclusions are variable, including dense gas, two-phase liquid vapor, and three-phase liquid vapor with daughter crystals (Prokofief et al., 2020). Nanoinclusions may contain extremely high concentrations of metal elements (Zhang et al., 2014; Prokofief et al., 2020), indicating that they can be regarded as an effective index for ore forecasting.

3.5. NPs in soil and water

The "clay fraction" in soil conventionally denotes a class of materials whose particles are smaller than 2 μm and show equivalent spherical diameters. Soil contains colloidal particles (1–1000 nm) including a part of NPs (<100 nm), which are usually of different sizes for the transport of nutrients, pollutants, organics, and hazardous elements (Navrotsky, 2003). Well-known examples of NPs in soil include aluminosilicate minerals; oxides and hydroxides of Al, Fe, and Mn; enzymes; humic substances; viruses; and mobile colloids (Kretzschmar and Schäfer, 2005). Organic NPs in
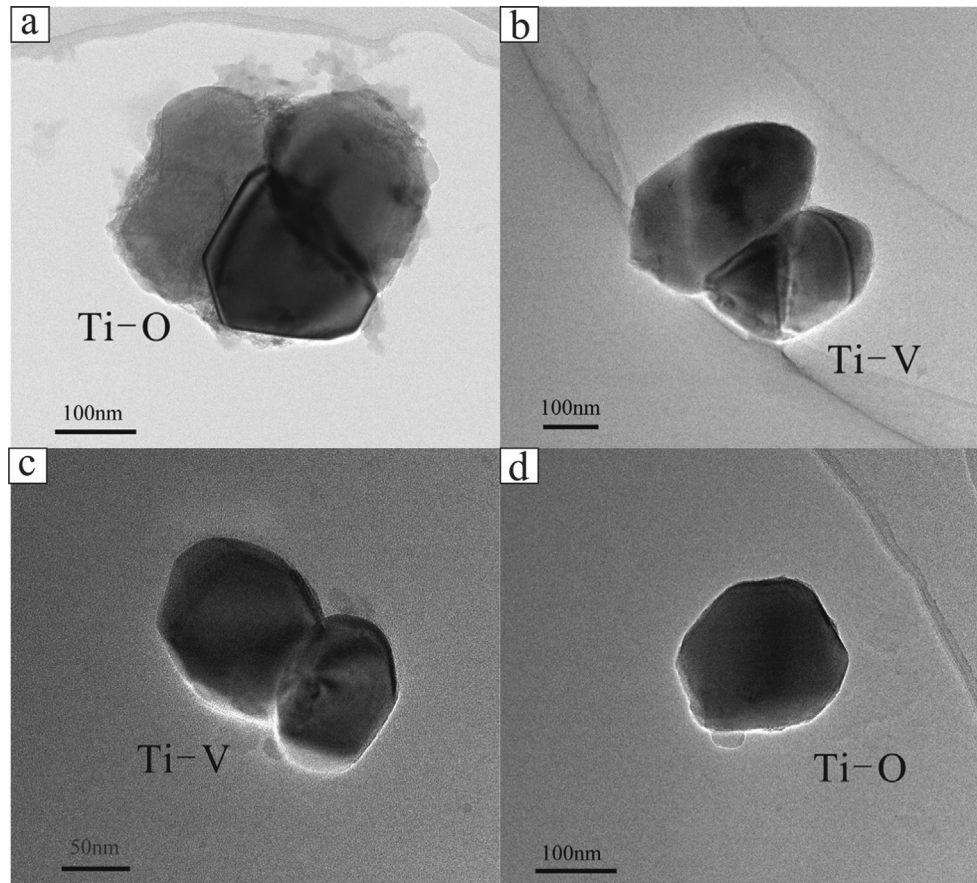

Fig. 3. TEM images of NPs containing Ti in the ascending geogas collected from the fault zone of a gold deposit in Henan Province, China, from Han et al. (2020): (A) Ti (65.34%), O (34.66%); (B) Ti (86.79%), V (3.76%), and Al; (C) Ti (91.18%), V (1.75%), and Al; and (D) Ti (64.66%) and O (32.37%).
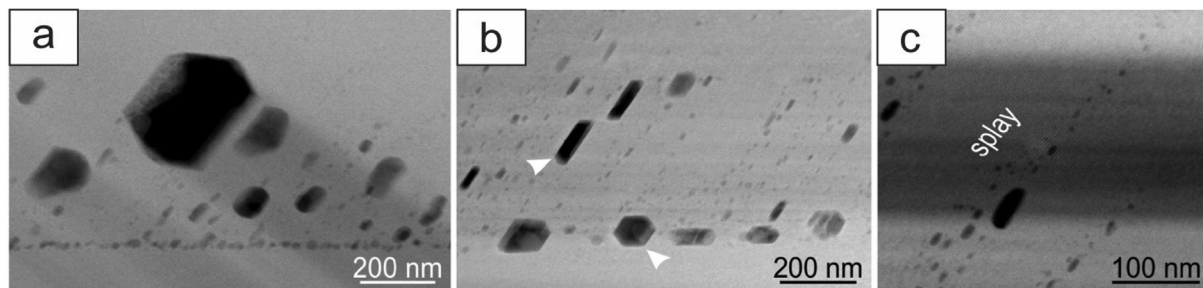
Fig. 4. HAADF STEM images showing the distribution of inclusions and cavities in hematite from Verdugo-Ihl et al. (2019): (a) finest inclusions (<20 nm) distributed as a continuous array; (b) mixture of inclusions and cavities at the nanoscale; and (c) details of the distribution of inclusions/cavities at the nanoscale.

soil are mostly associated with their inorganic counterparts or occur as coatings on mineral surfaces (Oades, 1989; Chorover et al., 2007). Because of their nature and surface properties, NPs in soil participate in essential ecological services, from regulating water storage and element cycling to sorbing and transporting chemical and biological contaminants, thus serving as a source or sink of organic carbon and plant nutrients. The formation (and transformation) of NPs in soil may be affected through an abiotic or a biological pathway or a combination of both. For example, clay minerals are largely formed by an abiotic pathways; humic substances are clearly biogenic because they represent the decomposition products of plant materials (biopolymers); and some Fe and Mn nanominerals in soil are formed through a combination of abiotic and biological pathways (León-Mejía et al., 2016; Sehn et al., 2016; Rodriguez-Iruretagoiena et al., 2016; Wilcox et al., 2015).

The emission of manmade or natural metal NPs can affect water quality, pollutant transformation, geochemical properties, and element cycles. Metal (oxide) NPs (Fig. 5) can enter the water environment in various ways (Liu et al., 2020b), where they threaten aquatic organisms, the ecological environment, and human health. NPs in water environments can regulate the transport of pollutants, affect the migration of metal elements, and reduce the content of pollutants and metal elements in sediments (Hochell, 2006; Hochella et al., 2008). The environmental behaviors of these NPs have attracted extensive attention in the environmental field in recent years (Georgin et al., 2021; Salomón et al., 2021; Streit et al., 2021). A large number of galena NPs are present in acid mine wastewater environments. The dissolution and release rate of galena NPs was 10–100 times faster than that of micron particles with the same properties (Liu et al., 2009a; Liu et al., 2009b). Thus, studying the characteristics of NPs in wastewater and their carrying and adsorption of other pollutants is highly significant for controlling water quality risks and ensuring water quality safety.

Due to their small size, colloids, particularly the nanosized colloids (one kind of NPs), have a much larger specific surface area than do larger particulates; moreover, the surfaces of many types of nanosized colloids contain a high proportion of functional binding groups for the adsorption of hazardous elements' adsorption. These colloids are at least as reactive as suspended particulates, can appear to be as mobile as dissolved solutes and are not subject to sedimentation in water (Sellaoui et al., 2021; Pereira et al., 2021; Zamberlan et al., 2020). This colloidal continuum can thus constitute an important carrier phase for metal transport in aquatic systems, a phenomenon described by the term nanovector (Hamon et al., 2005). Natural colloids consist of several important organic phases (e.g., humic substances and polysaccharides, as well as microbes) and inorganic phases (e.g., Fe and Mn oxides and aluminosilicates); of the latter, Fe oxides are among the most ubiquitous (Filella, 2007). Iron oxide and manganese oxide NPs are widely involved in the redox process of surface water and groundwater

because of their variable valence elements, which significantly affect the geochemical cycle of elements and the transformation of pollutants (Sehn et al., 2016; Wilcox et al., 2015; Rodriguez-Iruretagoiena et al., 2016). In recent years, research into NPs in the Earth surface in the fields of geology and environmental science has focused on interactions between NPs and environmental pollutants and their effects on pollutant migration and bioavailability (Oliveira et al., 2021b; Silva et al., 2021b; Sellaoui et al., 2021).

4. Origins of NPs related to geological processes in the Earth surface systems

Revealing the origin of NPs in the Earth surface systems is a prerequisite for gaining a deeper understanding of the geological processes in the crust and hazardous elements' migrations in soil and water. NPs are produced under various geological conditions and play an important role in the formation of macroscopic materials, understanding the formation of gemstones, the construction of Earth, the exploitation and use of natural metal NPs in ore deposits, and the treatment of hazardous elements in soil and water (Silva et al., 2021a, 2021b). Several major causes of natural NPs have been discussed and summarized (Hochella et al., 2008; Liu et al., 2008, 2009a; Waychunas and Zhang, 2008; Javadpour, 2009; Fathi and Akkutlu, 2013; Ju et al., 2016; Wang et al., 2016a, 2016b; Wang et al., 2016c; Zou et al., 2016). Understanding the origins of natural NPs in the Earth surface systems not only can provide helps for environmental geologists to know the sources of natural pollutant NPs, but also can indicate the information of concealed mineral resources for the ore deposit scientists. It must be noticed that the natural NPs in tailings or abandoned mines or other mined sites, which may be formed by means of mechanical or chemical or biological or the multigeological processes, can be transferred into pollutant NPs and may be damages to soil and water.

4.1. Physical origin

Some matters remain at the nanoscale during physical processes, such as weathering (Liu et al., 2018), fault activities (Zhang et al., 2019; Wang et al., 2019), and earthquake friction (Wang et al., 2016c). In a fault zone formed by sliding, rock can be crushed into NPs, and round and spherical NPs can vary and form lamellar structures and various tectonic patterns (Ju et al., 2017). In the process of earthquake fission, interactions between materials, such as high-speed shearing force and collisional friction, can generate NPs through thermal decomposition, abrasion, grinding, and powdering (Wang et al., 2016c; Ju et al., 2016). NPs of greater roundness and sphericity can be produced either by dynamic friction during brittle earthquakes or static friction during
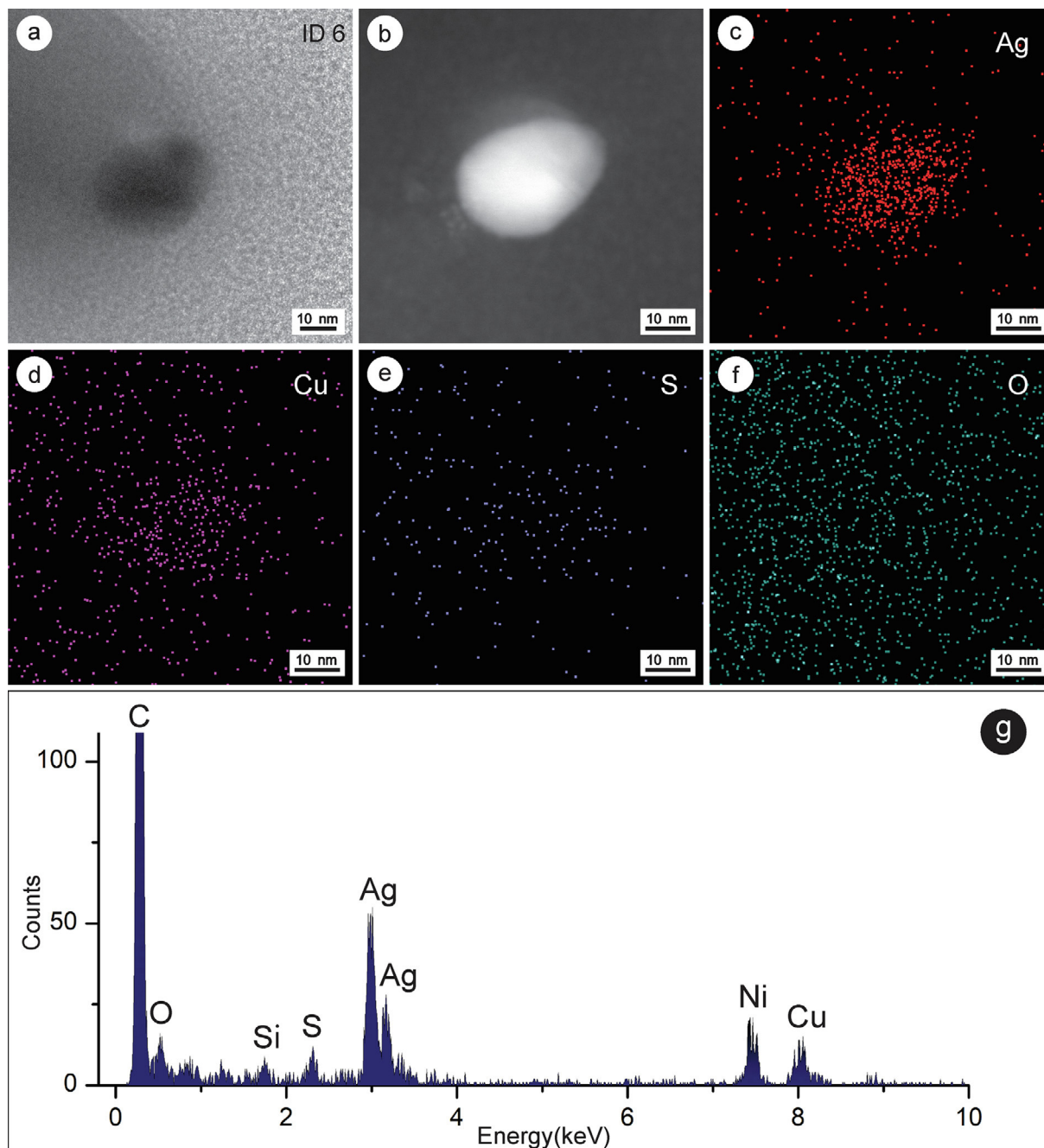
Fig. 5. NPs containing heavy metals, such as Ag and Cu, collected from the groundwater of gold ore, by Liu et al. (2020a): (a) TEM image; (b) STEM image; (c–f) elemental mapping images by EDS; and (g) EDS spectrum of particles in (a).

ductile earthquakes (Chao et al., 2009). The mechanical weathering of minerals is also an important formation process for NPs on the Earth surface (Hochella, 2002). Substances in the sea, lake, groundwater, hydrothermal fluid, and magma will separate and grow to form NPs under certain conditions (Theng and Yuan, 2008; Buseck and Adachi, 2008; Hochella et al., 2008; Wang et al., 2016a, 2016b; Ju et al., 2017; Yang et al., 2018).

4.2. Chemical origin

Chemical processes (mainly geochemical processes) provide an ideal environment for the formation of NPs. For example, element chemical reaction exists and concentration and temperature as well as pressure changes in the complex geochemical processes

(e.g., magmatism and hydrothermal processes), leading to that mineral crystallization or state transformation must go through the NP size stage. Some metal elements will form NPs and exist in the geological body (Zhang and Wang, 2018). This work summarized the main geochemical origins of NPs: (1) oxidation process origin. For example, Liu et al. (2020a) found NPs containing elements among the most abundant in the Earth's crust (O, Mg, K, Ca, Si, Al, Ti, Fe and P) and concealed ore body-related elements. Nearly all NPs were found to have O and S, indicating that these NPs were formed via oxidation of primary minerals in the concealed deposit; (2) precipitation process origin. The authigenic origin of native ore-bearing NPs is based on chemical transfer, which may be related to the precipitation of ore-forming matter from a colloidal solution (Wu et al., 2016; Wu et al., 2018; Yan et al.,

2018; Egglseder et al., 2019; Liu et al., 2019; Wierchowiec et al., 2021). For example, Wierchowiec et al. (2021) found the presence of nano native gold embedded in fine-grained assemblages of clay-ish masses and the absence of any evidence for abrasion of individual grains, suggesting the authigenic origin of native gold NPs. They proposed that these NPs were formed in situ through the chemical transfer relating to the precipitation of Au from a colloidal solution. McLeish et al (2021)'s research on the hyperenrichment of gold veins shows that the gold NPs flocculate and precipitate when the physicochemical conditions of an ore-forming hydrothermal solution (rich in Au element) change; (3) fluid-mineral interaction process. Using nanotechnological approaches, Verdugo-Ihl et al. (2019) reported that fluid–mineral interactions in the formation of copper-arsenic NPs can be fingerprinted at the atomic scale, where element exchange occurs. Wang et al. (2016a, 2016b) and Schindler et al. (2017) reported that fluids can mobilize U^{4+} under a reduction reaction in the form of uraninite NPs. Au has been shown to replace Fe in the structures of arsenopyrite and hydrothermal pyrite, thus leading to the formation of "invisible" Au (Trigub et al., 2017). Many amorphous NPs in different media can transfer ore-forming elements through fluid-mineral interaction processes and provide mineralization information (Liu et al., 2018); (4) The rapid crystallization process origin with both physical and chemical processes. For example, manganese nodules at the bottom of the ocean are cemented by NPs formed by the rapid cooling of mantle materials erupting from mid-ocean ridges (Zhang and Wang, 2018).

4.3. Microbial origin

Biological participation, particularly for the biomineralization which is defined by the mineral precipitation controlled or induced by microorganisms, protozoa, fungi, plants, and animals (Prozorov et al., 2013; Miot et al., 2014; Zan et al., 2016), also aids in the formation of NPs. Minerals formed by microorganisms are usually nanoscale, taking magnetosomes for instance, which are magnetic nanocrystals (magnetite or pyrite NPs according to Klumpp and Faivre (2012), Lin et al. (2017), and Lenagard et al. (2019)) wrapped in biofilm with a uniform size and single magnetic chip. Magnetosomes are formed under the effects of magnetotactic bacteria, which can control the process of crystal nucleation and growth using the protein in the cell, thus forming magnetite NPs that have a uniform size, shape, and chain arrangement; moreover, the iron oxide mineral particles produced by the extracellular respiration of iron-reducing bacteria often occur at the nanosize scale (Ding et al., 2010; Li et al., 2010; Wu et al., 2010b).

Moreover, with the rapid development of advanced research techniques as introduced in Section 2, more and more NPs related to the effects of biology were found. Ohnuki et al. (2015) found that the binding of Ce^{3+} to microbial surface functional groups can form Ce phosphate NPs. It was evidenced by experiments that pitchblende NPs can be formed by the reduction of bacterial enzyme (Suzuki et al., 2002; Min et al., 2005; Madden et al., 2012). Reith et al. (2016) found that biofilms can cause the precipitation of mobile nanoplatinum complexes and concluded that biofilms are capable of forming or transforming platinum-group nanomineral grains (Fig. 6) and may play an important role in platinum-group element dispersion and reconcentration in surface environments.

Different types of microorganisms are involved in the NP generation process induced by biomineralization. For example, sulfur oxidizing bacteria (Goffredi et al., 2004), metal oxidizing bacteria (Hallberg and Ferris, 2004), sulfur reducing bacteria (Labrenz et al., 2000), and metal reducing bacteria (Vetriani et al., 2005) participated in the generation of NPs induced by biomineralization in hydrothermal vents. Predecessors also discussed how proteins (such as cytochrome in Newsome et al. (2014)) and structure of bacteria (such as timbriae and flagella in Newsome et al. (2014)) promoted the biomineralization and the generation of NPs.

4.4. Multigeological process origin

Generally, NP formation, transformation, and deposition may be influenced by multiple geological processes, including the joint effects of geophysics, geochemistry, and geobiology. Taking gold and other metallic NPs as examples, NPs can form during the process of mineralization (Han et al., 2020; Hastie et al., 2020) and then be reformulated or reproduced by faulting and weathering (Han et al., 2020), the hydrothermal fluid-assisted liberation of Au (Hastie et al., 2020), or a migration process forced by groundwater (Langman and Moberly, 2018; Hu and Cao, 2019) or geogas (Jiang et al., 2019). Subsequently, aggregation and biological activity can change the NPs' status (Han et al., 2020). Both intrinsic and extrinsic factors drive the formation of gold NPs at fluid–mineral interfaces (Li et al., 2020b). In the Earth surface systems, many different natural rock/soil surfaces are covered by iron and manganese oxide mineral coatings that have nanolattice structures (Lu et al., 2019). The Mn-bearing minerals in the desert varnish can be regarded as an instance, which are extremely fine grained (typically < 100 nm) (Garvie et al., 2008) and are NPs. These Mn-bearing NPs are formed through repeated wetting and drying of the rock surfaces, evaporation and oxidation of soluble Mn^{2+}, and leaching and oxidation as well as precipitating of Mn (Garvie et al., 2008).

5. Properties of NPs in the Earth surface systems

The following sections discuss several major special properties of NPs in the Earth surface systems.

5.1. Size effect

Many studies have shown that NPs have different crystal structures, chemical activities, thermodynamic stabilities, and physical properties compared with the properties of micro- or macro-particles. When the particle is nanosized, a large proportion of atoms are located at the surface or near-surface environment. Thus, the special atomic arrangement and surface properties of nanomineral surfaces will significantly affect the properties of whole mineral particles, thus imparting different mineralogical properties compared with macrocrystals (Hochella et al., 2019). Significant differences in adsorption behavior (Ju et al., 2017; Luo et al., 2018), dissolution rate (Gilbert et al., 2004; Reich et al., 2005; Luo et al., 2008), thermal behavior (Reich et al., 2006), aggregation state (Wang et al., 2016a, 2016b), and catalytic activity occur between metal mineral NPs and their corresponding large-scale particles. For example, the melting points of gold NPs (2 nm in size) and nuggets are 327 °C and 1,064 °C, respectively (Buffat and Borel, 1976), while that of bulk copper and copper NPs are 1,083 °C and less than 490 °C, respectively (Liu et al., 2004). When the silver particle radius is less than 20 nm, the dissolution/melting temperature decreases rapidly with the particle radius (Luo et al., 2008).

5.2. Migration and aggregation effects

Natural metal NPs widely occur in soils, geogas, and ores in deposits, and such NPs tend to agglomerate into clustering forms (Wang et al., 2017b). Compared with macromineral crystals, these NPs easily agglomerate in the natural environment because of their small particle size and large surface energy. The agglomeration and migration of these NPs play a key role in ore-forming geological
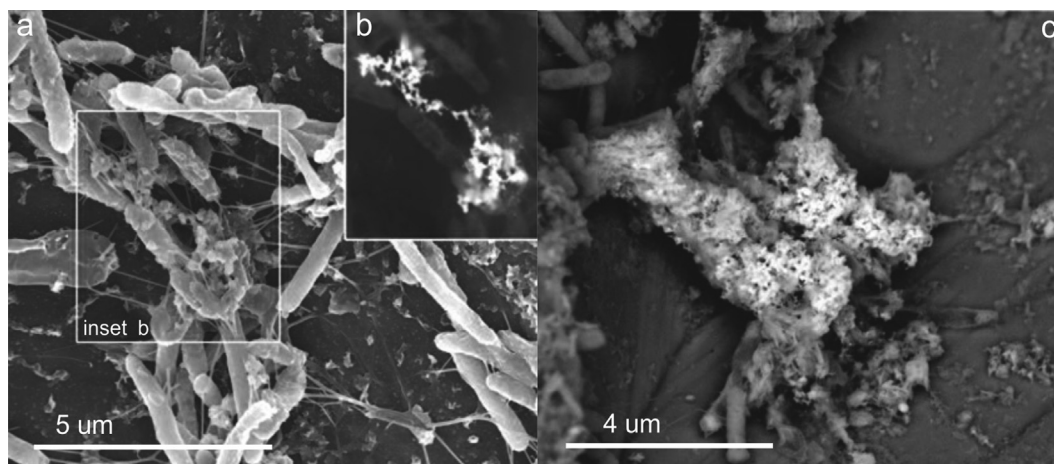
Fig. 6. Platinum biominerals formed by the bacterial activity of metalliduran biofilms from Reith et al. (2016): (a, b) SE and BSE images of Pt NPs associated with cells and exopolymeric biofilm substances, respectively; and (c) subsequent formation of larger Pt aggregates encapsulating biofilm sections (BSE image).

processes. In particular, NPs are easily dispersed in different media (gas, liquid, or solid) to form aerosol, colloidal, or solid solutions, which then experience long-distance migration (Wang et al., 2016a, 2016b). When microcracks are not developed, mineralization may occur (Ju et al., 2017). For example, colloform/botryoidal sphalerite aggregates that are optically anisotropic and have a strongly disordered structure or hexagonal habit are actually formed by the coalescence and agglomeration of colloidal nanocrystalline particles (Wu et al., 2018). Research on the distribution of gold NPs in the Beta Hunt gold deposit in Australia has shown that gold NPs can migrate stably in an ore-forming solution because they have the same charge on the surface as in hydrothermal physicochemical conditions (McLeish et al., 2021). The formation of irregularly shaped platy micronuggets of gold is attributed to agglomerations of isolated semispherical NPs (Wierchowiec et al., 2021), as documented by investigations of the growth history of individual native gold particles.

5.3. Adsorption effect

Many experiments have shown that under conditions of equal mass concentration, the adsorption capacity of NPs far exceeds that of their corresponding large-size particles. Sorption of gold NPs on pyrite may constitute a critical step in the metallogenesis process, with Luo et al. (2018) reporting that positively charged Au NPs could be significantly adsorbed on pyrite and that electrostatic interactions represent the dominant mechanism in the adsorption of Au NPs on pristine pyrite; moreover, by combining XPS data representing the Au spectra from adsorbed pyrite, they found that numerous Au NPs were adsorbed on the surface of pyrite at a pH of 2.2, whereas the number of adsorbed particles decreased with increases in pH (Fig. 7). The adsorption of Au and the formation of clusters of Au atoms can lead to the generation of NPs (Becker et al., 2010 and references therein; Reich et al., 2005, 2006, 2010). Mineral and amorphous particles can adsorb hazardous element ions in the environment by electrostatic forces, ion exchange, and surface complexation (Vieira et al., 2021). With decreases in particle size, the adsorption capacity and affinity for hazardous elements increase (Madden et al., 2006; Lead and Smith, 2009; Zeng et al., 2009; Barton et al., 2011; Yu et al., 2016) while the difficulty of desorbing hazardous element ions decreases (Wigginton et al., 2007). Mayo et al. (2007) reported that the arsenic adsorption efficiency of magnetite 12 nm in size is 200 times higher than that of magnetite particles at 300 nm in size. Accordingly, because of the

strong adsorption effect, NPs have great potential for use in removing of water pollutants.

5.4. Oxidation–reduction reactivity

NPs and large-scale mineral materials have significantly different redox activities (Sparks, 2010). NPs that have variable valence elements, such as iron oxide (Hough et al., 2011), manganese oxide (Zhmodik et al., 2012), and uranium oxide (Bonnetti et al., 2015; Schindler et al., 2017; Liu et al., 2018), are involved in the oxidation–reduction process in soil, sediment, surface water, and groundwater (Liu et al., 2018; Hu et al., 2018a; Hu and Cao, 2019) and have a significant effect on the geochemical cycle of elements and the formation and transformation of pollutants. NPs in the Earth surface soil and water can regulate the migration of pollutants, thus affecting the migration of metal elements and reducing their content in sediments (Hochella, 2006; Hochella et al., 2008). For example, sodium hydroxides, which are common in soil, can oxidize and degrade fungicides and bisphenol A as well as other emerging organic pollutants (Zhang et al., 2008; Lin et al., 2009). The large specific surface area and high surface energy of NPs can provide a large number of reactive sites for interfacial oxidation–reduction reactions (Liu et al., 2018). Taking natural gold NPs as an example, even different shaped NPs display unique chemical reactivity properties compared with bulk NPs, due to that surface area and reactivity vary significantly between gold NPs of different shapes (Hough et al., 2011). Iron ions in the magnetite lattice exist in a mixed valence state, which leads to high reducing activity. Vikesland et al. (2007) found that for the magnetite NP aggregation, the reduction activity rate decreases significantly as the size of magnetite NPs increases because the relatively smaller magnetite NPs have a relatively greater reactive surface area, better quantum confinement effects, and stronger diffusive availabilities.

6. Environmental effect of pollutant NPs and NPs used for soil and water remediation

Many NPs are found in the overburden of water and soil sediments on the Earth surface (Schindler and Hochella, 2016). These NPs include clay minerals that may be formed by weathering (Hochella, 2002) and contain a large number of metal NPs, especially hazardous element NPs (Singh and Lee, 2016; Gil-Diaz et al., 2017; Liang et al., 2017). Because of hazardous element NPs' influence on human health, researchers have paid particular
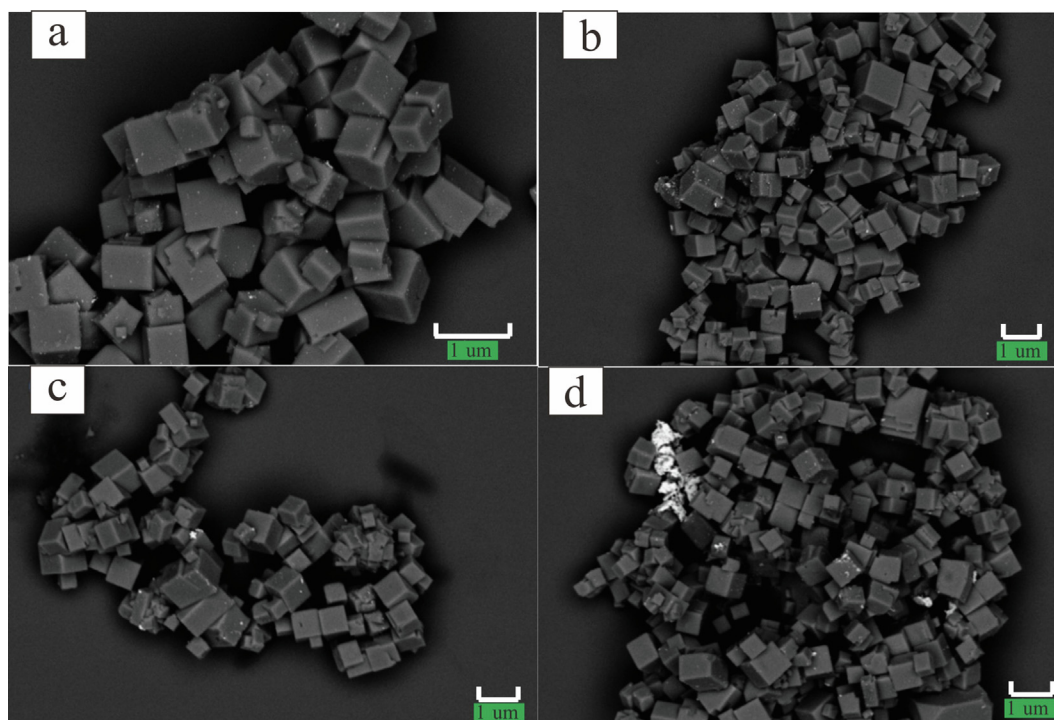
Fig. 7. SEM images of pyrite after sorption with positively charged Au NPs at different pH values from Luo et al. (2018): (a) pH = 2.2; (b) pH = 4.0; (c) pH = 7.1; and (d) pH = 10.0.

attention to the remediation and treatment of soil and water hazardous element pollution (Wu et al., 2010a; Crane and Scott, 2012; Hua et al., 2012; Bolan et al., 2014; Tesh and Scott, 2014; Yu et al., 2016; Zhao et al., 2016). Remediation technologies mainly include physical remediation, chemical remediation, phytoremediation, and biological (microbial) remediation (Lessen et al., 2008; Bolan et al., 2014). Because of their small size, large specific surface area, high surface activity, and strong reducing ability (Karn et al., 2009; Mukherjee et al., 2016; Ju et al., 2017; Su, 2017), NPs have played an increasingly important role in soil and water hazardous element pollution remediation (Crane and Scott, 2012; Hua et al., 2012; Tesh and Scott, 2014; Yu et al., 2016; Zhao et al., 2016; Arumugham et al., 2022; Xiao et al., 2022).

6.1. Environmental effect of pollutant NPs in soil and water

Soil and water pollution is serious in some areas of the world, particularly in areas where soil and water are affected by industry and mining. For example, the total over standard rate of soil in China is 16.1%, with over standard rates of Cd 7.0%, Hg 1.6%, As 2.7%, Cu 2.1%, Pb 1.5%, Cr 1.1%, Zn 0.9% and Ni 4.8% reported by the government in 2014. In some local areas, the heavy metal pollution of soils is serious; for example, Turhun et al. (2022) recently found that the average concentrations of As, Cd, Cr, and Hg in vineyard soils of the Turpan Basin exceed the background values of agricultural soils by factors of 1.05, 1.58, 1.49, and 1.15, respectively. Pollutant NPs, particularly for the engineered NPs can bring damage effects to environments such as soil and water.

NPs in soil mainly contain clay NPs, metal oxides NPs, and humic substance NPs (Theng and Yuan, 2008). NPs of soil can adsorb physically or combine chemically organic pollutants (such as polycyclic aromatic hydrocarbon) and inorganic pollutants (such as heavy metals, radioactive elements cesium and strontium, and compounds containing arsenic selenium and fluorine), forming pollutant NPs. Similarly, pollutant NPs in soil also have a wide

source range and distribution considering that the sources of pollutants in soil mainly include industrial and agricultural waste, chemical fertilizers, pesticides, atmospheric pollutant deposition, and accidental leakage. Hazardous element NPs in soil and water not only affect the yield and quality of crops and fish farming but also pass through the food chain and enter the human body, where they can damage human health. Accordingly, the remediation of soil and water contaminated by hazardous elements has always been one of the most important environmental issues addressed by environmental researchers (Wu et al., 2010a; Bolan et al., 2014).

Pollutant NPs in water primarily include carbon-based pollutant NPs, semiconductor pollutant NPs, metal and metal oxide pollutant NPs, and engineering polymer pollutant NPs (Bundschuh et al., 2018). Pollutant NPs have toxic effects on cells, microorganisms, aquatic organisms, and terrestrial organisms through the production of reactive oxygen species (Zhu et al., 2007), the release of toxic metal ions (Lubick, 2008), and the destruction of physiological and metabolic processes (Lyon et al., 2008). Predecessors reported that nano-sized zinc oxide (ZnO) and silver (Ag) will dissolve in water and produce toxic Zn^{2+} and Ag^+, which are adsorbed to the cell surface and block the transmission of the cell membrane, thus increasing the mortality of algae and mollusks (Król et al., 2017; Cornelis et al., 2017; Choi et al., 2018). Pollutant NPs have the following characteristics in water: (1) pollutant NPs have low mass concentration but high quantitative concentration in water, and they have wide source and distribution; (2) pollutant NPs in water have colloidal properties and can exist stably; (3) heavy metal elements in NPs may easily dissolve (e.g., carbonate NP bearing Pb, Zn, and Cd under acid condition, according to Huang et al. (2016) in water and lead to serious and rapid pollutions; (4) pollutant NPs have direct impacts on aquatic organisms, and even indirectly affect human life through biological amplification; (5) the final destination of heavy metal migration in water maybe the sediment; for example, the highest levels of 121 mg/kg Pb were found in the coastal sediments of the Arabian Sea along

the urban area of Pakistan, which was higher than the 49.5 mg/kg from surficial sediments of Lyari River (Waseem et al., 2014).

6.2. NPs for soil and water remediation

Due to the properties of large specific surface area and excellent catalytic activity, recently NPs have become a research hotspot in soil and water remediation. The followings discuss the NPs and modified NPs for soil and water remediation.

6.2.1. NPs remediation and evaluation of their effects

The primary NPs used in soil and water hazardous element pollution remediation include zero-valent iron–gold oxides, carbonaceous nanoparticle materials, polymer nanoparticle materials, and some modified nanoparticle materials. Fatimah et al. (2011) highlighted the pollutant methylene blue's removal efficiency with the uses of montmorillonite and the ZnO/montmorillonite. ZnO NPs in the latter were distributed in irregular sizes of about 2–10 nm. ZnO/montmorillonite and montmorillonite were found to achieve a 40% and 20% reduction of methylene blue after 150 min, respectively, because the surface area of the former is almost 4 times as large as that of the later. Similarly, results demonstrating that adding NPs can improve the removal efficiency of pollutants have also be found by Su et al. (2011) and Hashemian and Shaheli (2013). Lei et al. (2014) found that the maximum adsorption capacities of graphene oxide foam NPs in water are 252.5, 326.4, 381.3, and 587 mg/g for Cd^{2+}, Zn^{2+}, Pb^{2+} and Fe^{3+}, respectively, which are much higher than that of the most conventional adsorbents, indicating that graphene oxide foam NPs are ideal candidates for practical applications in the removal of a broad range of heavy metal ions.

The main mechanism underlying the remediation of soil and water polluted by hazardous element NPs is based on the unique characteristics of these NPs, such as their small particle size, large specific surface area, and high surface activity. In general, interactions between NPs and hazardous elements show very complex physical and chemical processes, with the main interaction mechanisms including adsorption, reduction, and oxidation (Braunschweig et al., 2013; Zou et al., 2016; Zhang et al., 2017). When evaluating the remediation effect of NPs on hazardous elements, such as soils, three types of indicators are typically used for assessment: soil property indices, which mainly include the pH value, hazardous element form, and the transformation of some extracted states (Ghrair et al., 2010); the effect of NPs on plants growing in hazardous element-contaminated soil; and the change index of hazardous element content in plants (Jin et al., 2016).

6.2.2. Modified NPs remediation for soil and water

Although some NPs, such as nanoscale zero-valent iron, have strong adsorption performance and reduction activity and can play a role in remediating hazardous elements in soil and water (El Awady et al., 2021), other NPs still have many shortcomings, including easy agglomeration and easy blunt as well as difficult recovery (Banerjee and Chen, 2007; Johnson et al., 2013; Zou et al., 2016). To decrease the NP agglomeration effect and improve the stability, reactivity, and mobility of NPs, researchers often modify NPs' material properties. The modification of NPs introduces specific functional groups or combines certain substances that have special properties when preparing NPs with the purpose of creating NPs with new characteristics and improving their effect on pollutants. Methods for modifying NPs usually include coating, loading, mixing, and surface changing (Lin et al., 2010; Johnson et al., 2013; Chen et al., 2017).

The remediation effect of hazardous elements in soil and water can be improved by combining NPs by increasing the soil pH and adsorption sites, as well as stability (Li et al., 2012; Luo et al.,

2013). Reports have shown that the Pb^{2+} removal capability in an aqueous solution can be enhanced by the application of Mg $(OH)_2$-supported nanoscale zero valent iron (Fig. 8) (Liu et al., 2015), and that the application of nanocomposite materials when remediating chromium pollution in acidic soil can effectively improve the soil's pH and, reduce the mobility of chromium, thereby significantly reducing the uptake of chromium by maize (Wang et al., 2017a).

6.3. Remediation technology of nanomaterial combination

To improve the remediating effects, NPs can be used in combination with other remediation technologies. Two main typical nanomaterial combination techniques are presented below.

6.3.1. Combination of NPs and electrical remediation techniques

In this combined technology, NPs adsorb hazardous element ions and enhance the transport potential of NPs through electrophoresis; thus, these two technologies are exploited and the remediation of soil and water hazardous element pollution is improved (Lima et al., 2017). Studies have found that electric processes can promote the diffusion and migration of NPs, such as nano-iron in coarse and medium granular soils (Chowdhury et al., 2012), and they have also shown that electrodialysis in the electrokinetic process in clay soil can increase the diffusion and migration of nano-iron (Gomes et al., 2013). Electrical remediation and permeable reactive barrier combination technology can reduce 88% of Cr^{6+} in Cr-polluted soil (Shariatmadari et al., 2009). Herrada et al. (2014) introduced the combination technology of electrokinetic remediation and nano-iron as a catalyst for in-situ treatment of pollutants in soil. They found that nano-iron can significantly enhance the efficiency of electrokinetic remediation, because nano-iron as an electrocatalyst enhances the kinetic process of the reaction. The sliver modified fullerene/layered double hydroxides composite (C_{60}@AgCl-LDO) NPs are created for photoelectrically degrading Bisphenol A in water. The Bisphenol A degradation by irradiation using simulated visible-light was 99% for 5 cycles, when C_{60}@AgCl-LDO was used as the photoelectric catalyst in 0.1 mol/L NaCl electrolyte with an applied voltage of −1.0 eV for 2 h (Ju et al., 2021).

6.3.2. Combination of NPs and phytoremediation techniques

In this combined remediation technology, NPs improve plants' ability to uptake hazardous elements by reducing hazardous element toxicity to plants. Singh and Lee (2016) found that the addition of nanoTiO$_2$ to soil can promote the uptake of Cd by soybeans (Fig. 9), with nanoTiO$_2$ particles entering the chloroplasts of soybean plant tissues and then combining with the reaction center of photosystem II (Fig. 9) to improve the electron transport and photoadaptation ability of chloroplasts, thereby facilitating the soybean absorption of Cd to improve phytoremediation. Lou et al. (2016) reported that the combined use of Fe$_2$O$_3$ NPs, alfalfa, and Rhizobium can effectively reduce the total Cd content in soil and the Cd content of aboveground edible parts of alfalfa. Such a combination is an effective two-way remediation system. In addition, when combining NPs technology and phytoremediation, various NPs and application times will affect the remediation capacity (Wang et al., 2014b; Liang et al., 2017).

6.4. Main factors influencing the remediation of NPs

The application of NPs for soil and water hazardous element remediation is affected by many factors, primarily the physical and chemical characteristics of the soil and water, the different hazardous elements, and the NPs themselves.
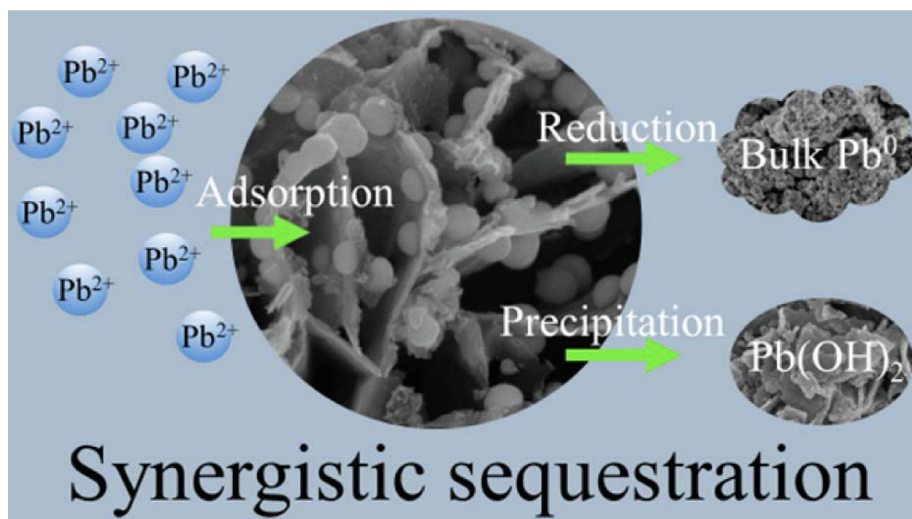
Fig. 8. Three paths are involved in the synergistic removal process of Pb^{2+} from an aqueous solution according to Liu et al. (2015): (1) Pb^{2+} is adsorbed by $Mg(OH)_2$ (accompanied by ion exchange reaction); (2) Pb^{2+} is reduced to Pb by nanoscale zero valent iron, and (3) Pb^{2+} is precipitated as $Pb(OH)_2$. The hydroxies provided by $Mg(OH)_2$ can dramatically promote the role of nanoscale zero valent iron as a reducer, thereby greatly enhancing the entire Pb^{2+} sequestration process.
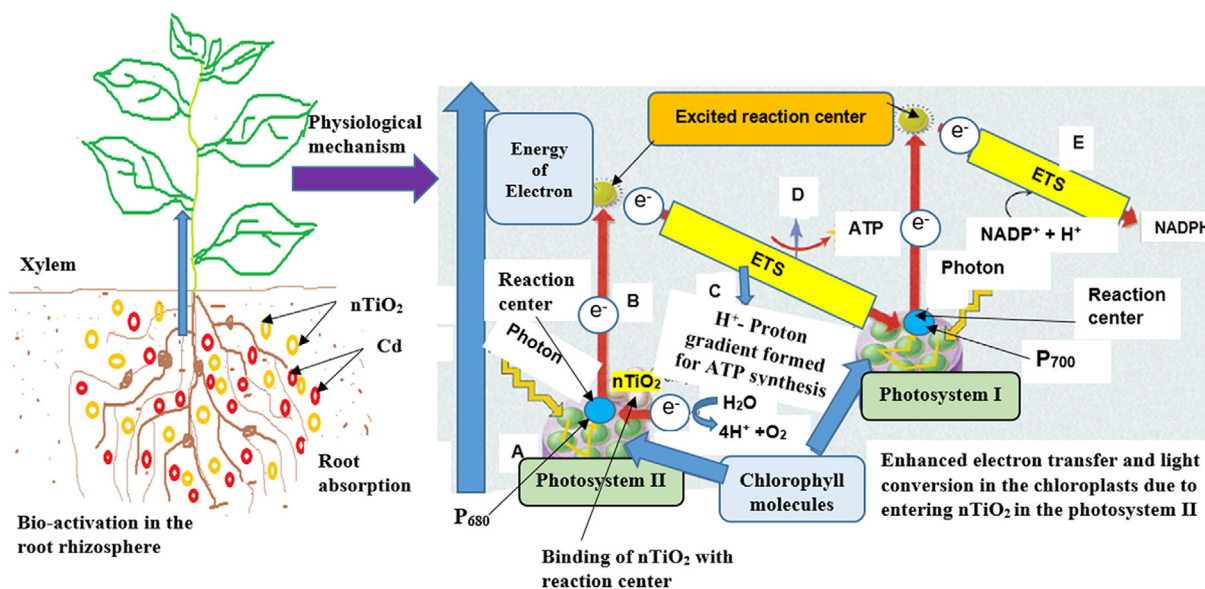

Fig. 9. Possible mechanism of Cd accumulation in soybean plants in the presence of nanoTiO$_2$ particles in soil, from Singh and Lee (2016).

6.4.1. Physical and chemical characters of soil and water

The remediation of hazardous elements in soil and water is affected by many factors, such as soil organic matter, iron and aluminum oxides, clay content, and soil pH (Eggleton and Thomas, 2004; Kim et al., 2015). For example, organic matter and iron and aluminum oxides in the soil interact with NPs, which influences the remediation effect of NPs on hazardous elements in soil (Vítková et al., 2015).

Soil pH is a particularly sensitive indicator of the bioavailability of hazardous elements in soil (Vítková et al., 2017). Liu et al. (2017) reported that after the addition of certain NPs, the value of soil pH increased, which reduced the bioavailability of hazardous elements (Fig. 10a), leading to good remediation effect. In a field experiment, the application of nanosubmicron mineral-based soil modifiers to soil increased the soil pH from 5.90 to 6.07, which reduced the

uptake of Cd by rice. However, some studies have found that adding NPs can reduce soil pH (e.g., the use of valent iron–copper bimetal (nZVI/Cu) NP can induce the generation of H^+ during the removal of hexavalent chromium in soils, according to Zhu et al. (2016) because the nZVI/Cu react with OH^-, inducing more H^+ and affecting the pH of soils). For example, adding modified nanocarbon to Cu- and Zn-contaminated soil reduces the soil pH by 0.5–1.0 units, which significantly hinders the remediation of hazardous elements in soil (Wang et al., 2009). Some studies have focused on reducing soil pH and changing the hazardous element forms by employing NPs to reduce toxicity (Zhu et al., 2016). In addition, soil redox conditions may affect the effects of nanomaterials. Under normal circumstances, the application of NPs can cause nanoiron oxide to adsorb harmful element pollutants and form coprecipitation. In addition, the condition is also influenced by
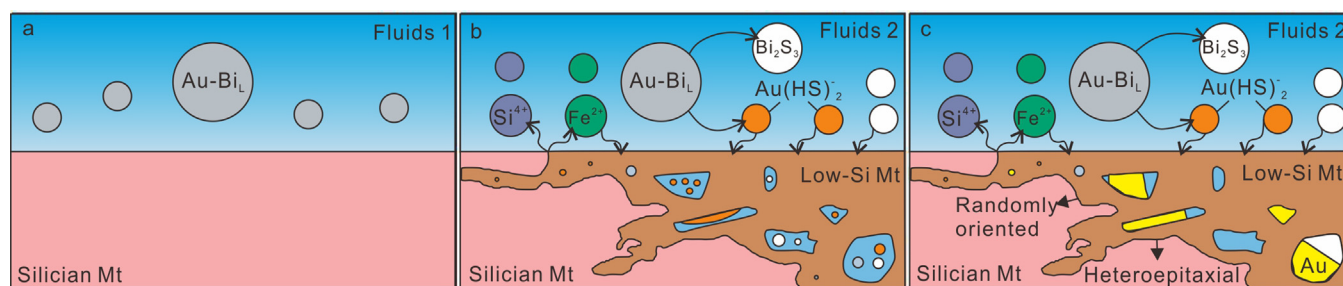
Fig. 10. Gold nanoparticle formation at mineral–fluid interfaces from Zhou et al. (2021): (a) crystallization of silician magnetite from fluids that carried nanosized Au–Bi melts; (b) coupled dissolution–reprecipitation reaction on silician magnetite surfaces forming porous low-Si magnetite and causing the disequilibrium of the Au–Bi melts that formed Au-supersaturated fluids at the pore scale; and (c) the heteroepitaxial and randomly oriented growth of gold NPs in the nanopores of low-Si magnetite.

other factors, such as the amount of NP material applied, the type of soil and the exposure time of the soil (Fajardo et al., 2015; Gil-Diaz et al., 2017).

The pH, ionic strength, natural organic matter and clay in water can influence the water remediation effect using NPs. Guzman et al. (2006) reported that pH of water can influence the aggregation of titania NPs, and the less aggregation, the better water remediation effect. Petosa et al. (2012) found that the polymer-coated NP suspensions are stable over a large range of $NaNO_3$ concentrations to achieve good remediation effect. Thio et al. (2011) demonstrated that natural organic matter drastically increased the stability of TiO_2 NPs in water under most conditions, due to the combined effect of increased electrostatic and steric repulsions. The stability TiO_2 NPs in water benefits the photocatalytic reaction and can lead to good remediation result. The clay in water can affect the suspension stability of NPs such as carbon nanotubes and further influence the remediation effects (Han et al., 2008).

6.4.2. Hazardous element species

The removal or stabilization effects of various hazardous elements using the same NPs are usually different. For example, Wang et al. (2009) used modified nanocarbon black to reduce available Cu and Zn in soil and found that modified nanocarbon black reduced available Cu in soil far better than Zn. Gil-Diaz et al. (2017) also found that the application of iron NPs reduced the bioavailability of As and Hg in soil to different degrees. Iron NPs showed better results for As immobilization, compared with Hg. Lei et al. (2014) found that the maximum adsorption capacities of graphene oxide foam NPs in water for different heavy metal ions are different, as demonstrated in Section 6.2.1, leading to different remediation effects.

6.4.3. Influence of NPs themselves

NPs have relative greater specific surface area, stronger adsorption and chelation ability, and better catalytic activity, which makes NP remediation technology overcome some shortcomings of traditional remediation technology and shows high remediation efficiency in the remediation of contaminated soil and water. However, some NPs easily agglomerate and oxidize and can lead to bad remediation effects. For instance, the exposed nanoscale zero-valent iron (nZVI) is easy to agglomerate and be oxidized by water or dissolved oxygen in the medium to form a passivation layer, resulting in the rapid reduction of its reaction activity and migration ability in the soil, making it is difficult to degrade the target organic pollutants (Stefaniuk et al., 2016).

Various NPs have different remediative effects on hazardous elements in soil and water. Mallampati et al. (2013) applied different nanofenanthrenes to soils contaminated with Cd and found that these NPs could fix 95%~99% of the hazardous elements in

the soil, with different NPs showing different effects. This is because the reduction capability and adsorption ability as well as precipitation degree of heavy metals differ by NP material types. Wu et al. (2012) reported that organo-montmorillonite supported iron NPs were found to be more efficient in the removal of Cr^{6+} in water than unsupported iron NPs. The reaction ratio was accelerated by an increase in acidity and iron loading. They also found that the Cr^{6+} removal rate using organo-montmorillonite supported iron NPs is very fast during the very first 0–20 min of reaction time, and the removal rate of the subsequent 20–350 min was very slow.

The application of different quantities of NPs can also lead to different repair effects among hazardous elements in soil. The main NPs that can be useful for remediation include metal NPs, carbon-based NPs, and polymeric NPs. In general, the more NPs applied to the soil, the better the hazardous element remediation effect (Zhu et al., 2016), but when the dosage increases to a certain extent, the effect will not increase. For instance, Ahmed et al. (2021) showed that when the adsorbent (a novel organic clay) dosage was increased, the adsorption efficiency of As^{3+} increased and the adsorption capacity for As^{3+} reached 99% approximately at the $6 \ g \cdot L^{-1}$ dose. In addition, the interaction times of NPs and soils also affect the restoration effect of hazardous elements in the soil. Generally, longer interaction times produce better effects (Liang et al., 2017).

NPs used in the remediation of polluted soil can not only ease the pollution, but also can be used for the growth of plants. Lin et al. (2020) reported that nZVI also has the potential to be used as nano fertilizer and iron supplement, and an appropriate concentration can promote the growth of plants. However, at present, there is a lack of research in this field, and the relevant mechanism and legal rules for guiding its use must be further explored, though legal rules guiding the use of engineered nanomaterial have been made and exerted by many countries such as USA, UK, Japan, European Union, and China.

7. Resource effect of NPs and their use for the resources exploration

NPs in the Earth surface systems are closely related to the formation of resources such as ore deposits and hydrocarbon reservoirs. The resource effect of NPs and how NPs can indicate the resource exploration are introduced in this section.

7.1. Resource effect of NPs

Nanomineralization in the Earth surface generally includes submarine hydrothermal sedimentary mineralization, nanobiominer-

alization, weathered clay deposits, lateritic rare earth deposits, and convergence mineralization induced by NPs in underground gases. However, most ore deposits related to the mineralization process are concealed. The special geophysical and geochemical properties of NPs, such as high surface energy, high redox activity, strong adsorption, easy migration and aggregation, as reviewed in the preceding section, allow ore-forming elements to enter the ore-forming fluid more easily through micro- or nano-channel and fractures of rocks. NPs can also be transported by ion diffusion, electrochemistry, and flow of underground gases. The formation, migration, deposition, and accumulation of NPs are closely related to the resource enrichment and evolution, and therefore, NPs have resource effects and can be applied to resource exploration.

Ore-bearing NPs that originate with concealed ore bodies, can migrate via hydrothermal fluids and crystallize from supersaturated fluids (Zhou et al., 2021). They can also be carried or transported by groundwater flow (Liu et al., 2018; Yi et al., 2019; Liu et al., 2020a; Cao and Chen, 2020) and ascending gas flow (Tong et al., 2002; Wang et al., 2012; Ye et al., 2014; Liu et al., 2018; Jiang et al., 2019; Han et al., 2020) as well as winds (Wang et al., 2016c), and finally discharge to the catchment basin (Yi et al., 2019) and then deposit in soil (Luo et al., 2015; Zhang et al., 2015; Liu et al., 2018) or accumulate in plants or animals (Hu et al., 2017a, 2018a). These ore-bearing NPs, which contain various degrees of ore-related elements (e.g., Ag, Cu, Zn, La, Ce, Pb, and Pt), are rarely found in natural settings unrelated to ore bodies or mineral extraction (Liu et al., 2020a) and can enrich mineralization or even reach the surface under appropriate redox conditions or in an appropriate space. Accordingly, NPs in geogas, groundwater, soil, or plants and animals can be regarded as indicators and used to guide the exploration of these concealed deposits (Lu et al., 2021; Wang et al., 2021).

7.2. NPs of hydrothermal origin: Indicating the mineralizing

Ore-bearing NPs transformed by hydrothermal fluids have been widely studied to reveal the mechanisms of mineral crystal formation, growth, and transformation so that the prospecting criteria for ore exploration can be proposed. Taking hydrothermally generated invisible gold NPs as an example, reservoirs of fluids that contain gold NPs in Earth's lithosphere can be a source of Au-bearing fluids for the formation of gold deposits (Prokofief et al., 2020). Hough et al. (2011) reported that gold NPs are formed by direct precipitation from a hydrothermal fluid or exsolved from an arsenian pyrite matrix during the later stages of a deposit's evolution, and they can be transported upward during deformation and metamorphism (Hastie et al., 2020) and then finally deposited due to fluid immiscibility (Zhu, 1996), cooling and/or boiling (Hough et al., 2011), or trapped by pore spaces created by crystal plastic deformation (Li et al., 2020b). The transport and deposition of gold from colloidal suspensions in hydrothermal fluids has been a persistent theme in ore deposit research (Hannington et al., 2019).

Significant amounts of minor and trace elements, including As, Pb, Sb, Bi, Cu, Co, Ni, Au, Zn, Ag, Se, and Te, can be incorporated into NPs (Filimonova and Trubkin, 2008; Deditius et al., 2011). These elements can be transferred into supersaturated hydrothermal fluids. For example, the crystallization (Fig. 10a) and dissolution–reprecipitation (Fig. 10b) processes of gold-bearing NPs can aid in the formation of Au-supersaturated fluids, which crystallize and grow in different ways, e.g., directionally or randomly (Fig. 10c), depending on the intrinsic crystal structure and pore textures (Zhou et al., 2021). Although the ore content is sometimes low in ore-bearing formations because of low crystallization temperatures, later fluid activity may contribute to ore enrichment (Zhang et al., 2015).

7.3. Ore-bearing NPs: Concealed deposit indicator

Ore-bearing NPs can be regarded as concealed deposit indicators; for example, numerous metal-bearing NPs have been observed in geogas (Cao et al., 2009, 2010a, 2010b, 2017; Wang et al., 2016a, 2016b, 2017b; Wei et al., 2013; Zhang et al., 2015). Geogas prospecting is a potential method for exploring concealed ore bodies because ore-bearing NPs can be transported by geogas (Jiang et al., 2019). Recently, Jiang et al. (2019) and Han et al. (2020) indicated that gold and other natural metal NPs can be generated by faulting and weathering based on previous mineralization, with more metallic NPs observed in geogas collected in the fault zone, thus leading to easy capture and ascension by means of gas flows (Fig. 11). Natural metal NPs can be adsorbed onto the surface of gas bubbles and migrate upward with the bubbles (Fig. 12) (Wang et al., 2017b; Zhang et al., 2019). The forces that drive NP transportation may include temperature and pressure differences and the Earth's magnetic field (Han et al., 2020), and the particle migration environment should be oxidative.

Metallic NPs in deep fault gouges can migrate farther into surface soils or water (Zhang et al., 2019). Liu et al. (2020a) classified NPs in groundwater as native metal NPs, metal-based NPs, and carrier NPs associated with trace elements (Fig. 13) and proposed that these NPs enable the efficient transport of ore-forming elements in groundwater and reflect the properties of deep concealed ores because the metal-bearing NPs in groundwater are similar in composition to deep ores (Cheng et al., 2018; Hu and Cao, 2019; Yi et al., 2019; Cao and Chen, 2020). Additionally, because of the special properties of NPs, metal-bearing NPs can facilitate the transportation of metals in groundwater (Yi et al., 2019); moreover, the NP form is the dominant transportation form of ore-forming substances (Fig. 13). Fig. 14 depicts Ag-related NP formation and transportation as well as deposition processes, indicating that in thick overburdened areas, where local geochemical information on abnormal elemental levels is not obvious and geophysical prospecting methods may have multiple solutions, the accuracy of prospecting can be improved by using NPs in groundwater that provide concealed ore body information (Cheng et al., 2018; Hu and Cao, 2019; Yi et al., 2019; Cao and Chen, 2020).

Geochemical explorations of concealed deposits covered by thick rocks and soils present special problems (Wang et al., 2017b). Native gold and Hg-, Cu-, Zn-, Pb-, and W-bearing NPs can be found in the surface soil in concealed deposits (Zhang et al., 2019) because gases and other particles are trapped by soil geochemical barriers (Wang et al., 2017b), indicating that NPs in the soil come from ore bodies because they show the same features (Lu et al., 2017). The NPs of Au and Cu and ion complexes of U are more readily absorbed into fine fractions of soils containing clays, colloids, oxides, and organic matter (Wang et al., 2016a, 2016b; Liu et al., 2020a). Higher concentrations of trace elements in soils from the mining area result from the vertical migration of ore-forming elements with geogas or groundwater after mineralization processes and very high concentrations of ore-forming elements in the fault gouge (Yi et al., 2019; Zhang et al., 2019).

7.4. Organic NPs: Indicators of unconventional reservoir formation and mineralization

Research into unconventional gas and oil reservoirs is based on a new understanding of the massive storage capacity of organic NPs and nanopores in tight reservoirs (Zou et al., 2013). Although tight reservoirs are continuously or quasi-continuously distributed (Zou et al., 2013; Gonciaruk et al., 2021), organic NPs and nanopores are dispersed along with inorganic matter, such as clay minerals (Loucks et al., 2012; Zhao et al., 2019). Nanopores in a tight reservoir, such as shales, are widely developed and can not only
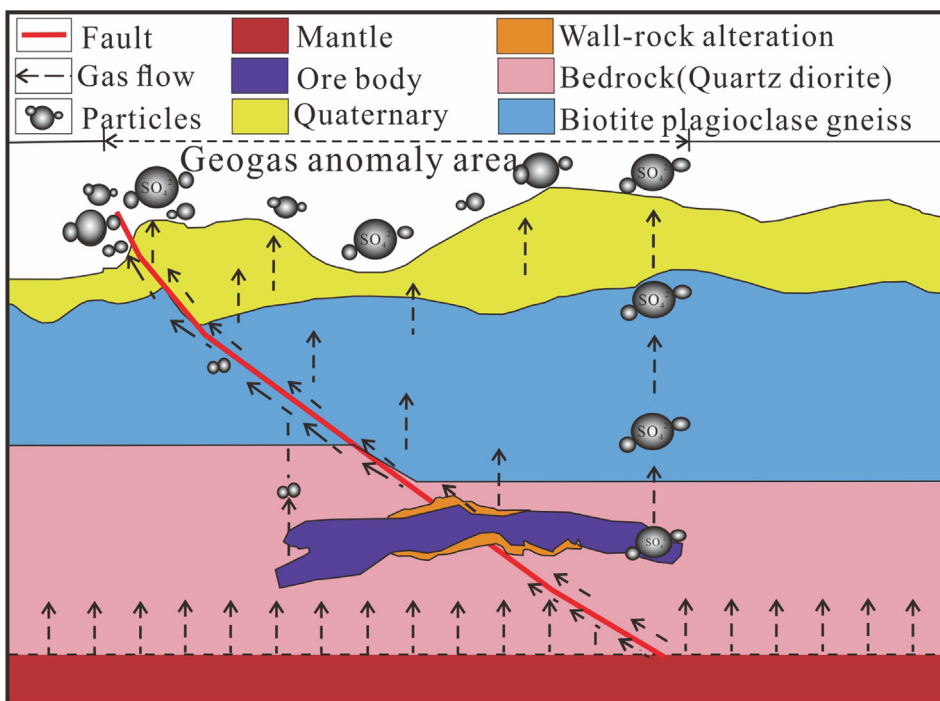
Fig. 11. Particle migration by ascending gas flow, which was modified from Jiang et al. (2019).
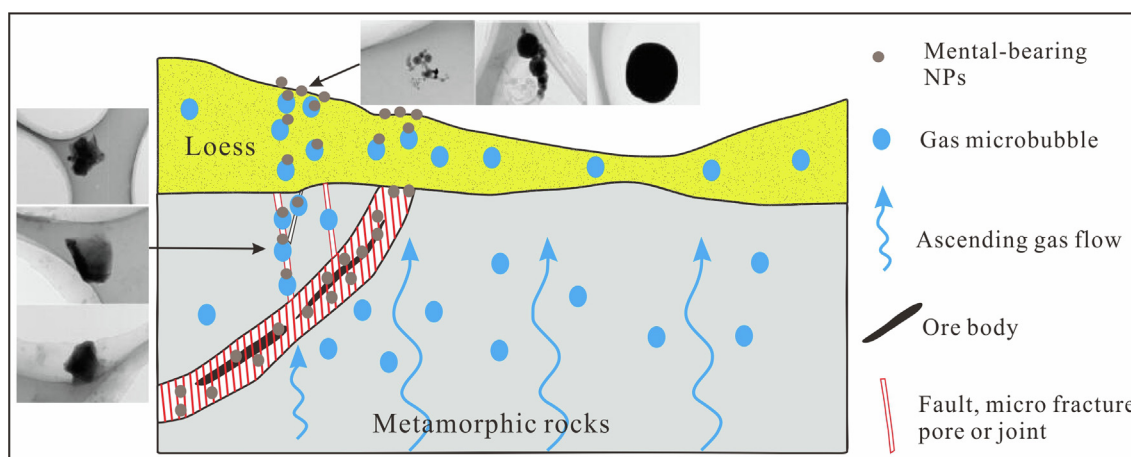

Fig. 12. Metal-bearing NP migration model under the function of ascending geogas, which was modified from Zhang et al. (2019).

reflect the hydrocarbon-generating stages (Modica et al., 2012; Mastalerz et al., 2013) but also determine the gas and oil occurrence states (Hu et al., 2015a). Nanopores' evolution are complicated by internal and external factors (Clarkson et al., 2016). Compared with micro- or macroscale organic matter (kerogen), organic NPs may have different hydrocarbon generation characteristics and models (Wang et al., 2014a). In particular, nanopores located in NPs may be more complex because of their nanoproperties.

Currently, an understanding of the nanoeffects and their corresponding influences on organic NPs hydrocarbon generation remains incomplete. For example, the deformation, hydrocarbon generation speed, and pore space variation mechanisms of organic NPs are still not well understood, and the process and mechanism of interactions between NPs and inorganic matter must be further investigated. To improve the efficiency of exploration and development, the generation, adsorption, diffusion, migration, and occur-

rence of unconventional gas and oil must be revealed by solving these scientific problems.

Organic matter and various organic fluids are also widely involved in the mineralization process of metal elements, as confirmed by numerous researchers (Saxby, 1976; Large et al., 2007; Fuchs et al., 2015; Chen et al., 2015). Sedimentary organic strata (coal, shale, and carbonate rocks) are rich in nanoorganic pore structures (Ju et al., 2017; Lu et al., 2017), and they can provide reduction and adsorption barriers to accelerate mineralization processes, such as Carlin-type gold deposits (Sililtoe and Bonham, 1990; Hulen et al., 1998; Hu et al., 2002; Su et al., 2008; Muntean et al., 2011; Chen et al., 2015; Ma et al., 2017), orogenic gold deposits hosted by metamorphic sedimentary rocks (Hu et al., 2015b, 2017b; Chen et al., 2018), MVT lead–zinc deposits (Leach et al., 2010, 2017; Song et al., 2019, 2020), sandstone- or shale-type copper–uranium deposits (Cathles and Adams, 2005; Brown, 2006; Hitzman et al., 2010; Hayes et al., 2012), metallic sul-
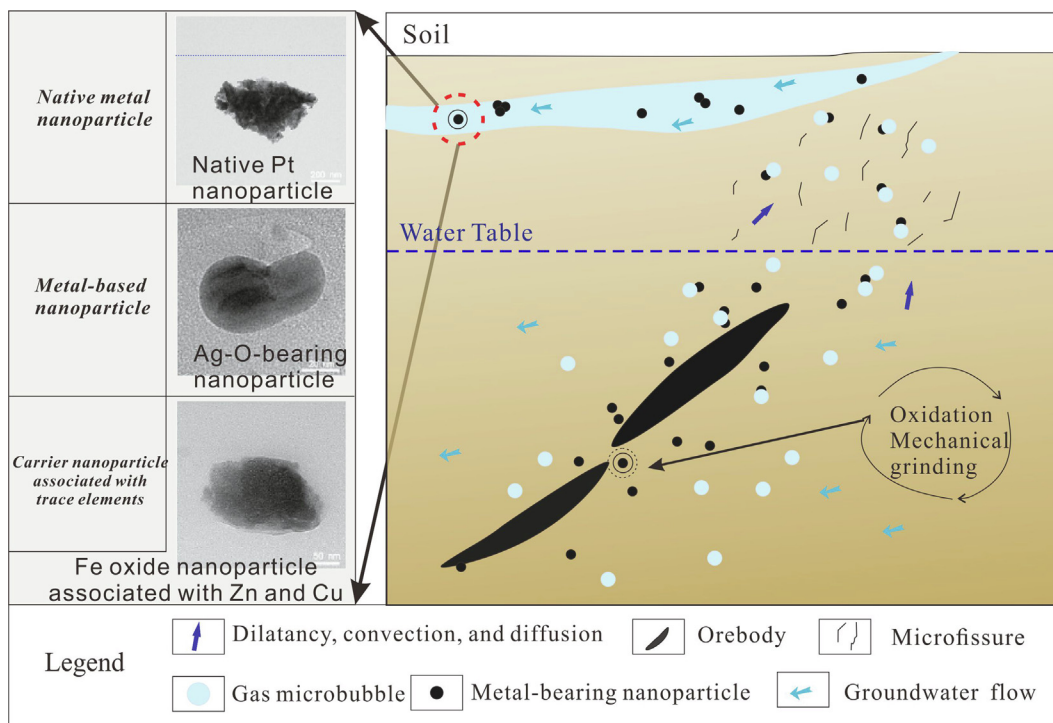
Fig. 13. Ore-related NP formation and transport, which was modified from Liu et al. (2020a). Three main types of NPs participate in the transport of ore-forming elements in groundwater: native metal NPs, metal-based NPs, and carrier NPs associated with trace elements.
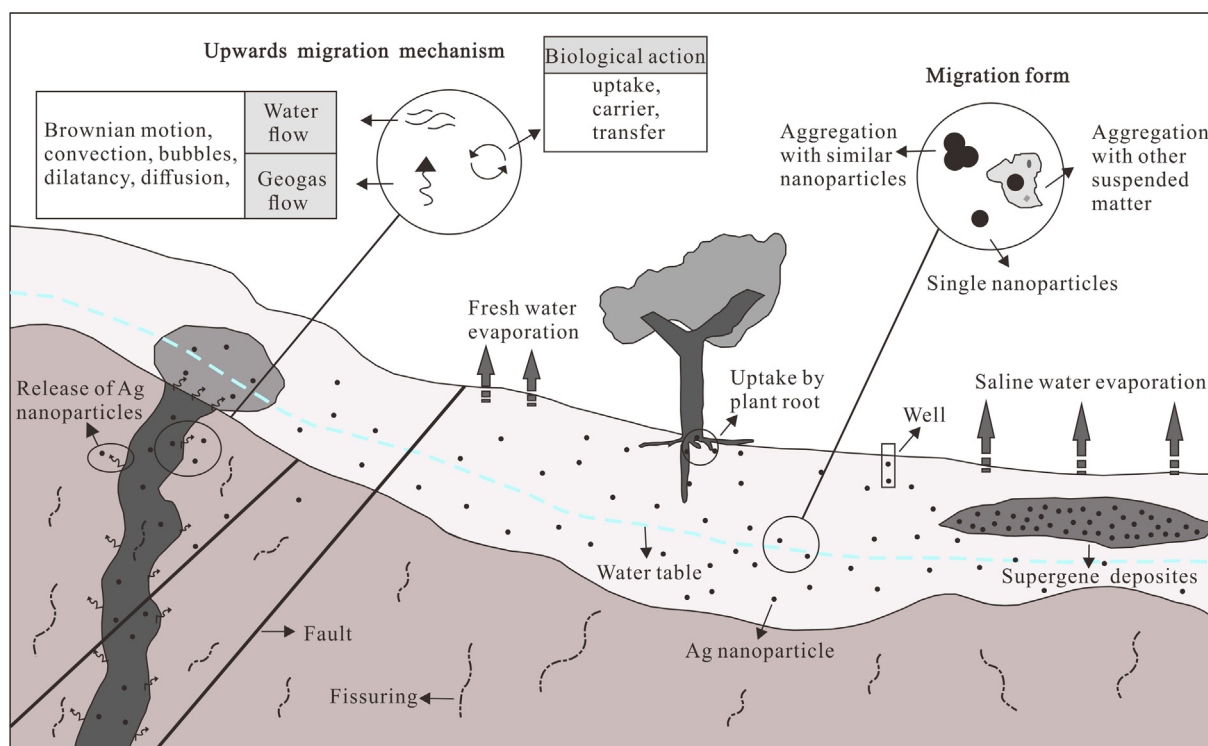

Fig. 14. Processes of supergene silver deposits formed by Ag NPs in a groundwater system, modified from Hu and Cao (2019).

fide deposits in black shale (Pagès et al., 2018), polymetallic deposits hosted by sedimentary rocks (Hu et al., 2002), and coal-type rare metal deposits (Hu et al., 1999; Du et al., 2009; Lewińska-Preis et al., 2009; Seredin, 2012; Seredin and Dai, 2012; Dai et al., 2018). Previous researchers have intensively investigated the characteristics of and controlling factors behind the coexistence of organic matter and metal deposits (Fig. 15). However, the fluid migration and interaction of organic matter and metal deposits at the nanoscale have not been revealed. The interaction mechanisms and coexistence models between organic matter and metal deposits in different typical sedimentary basins on multiple scales (particularly the nanoscale) require further study.
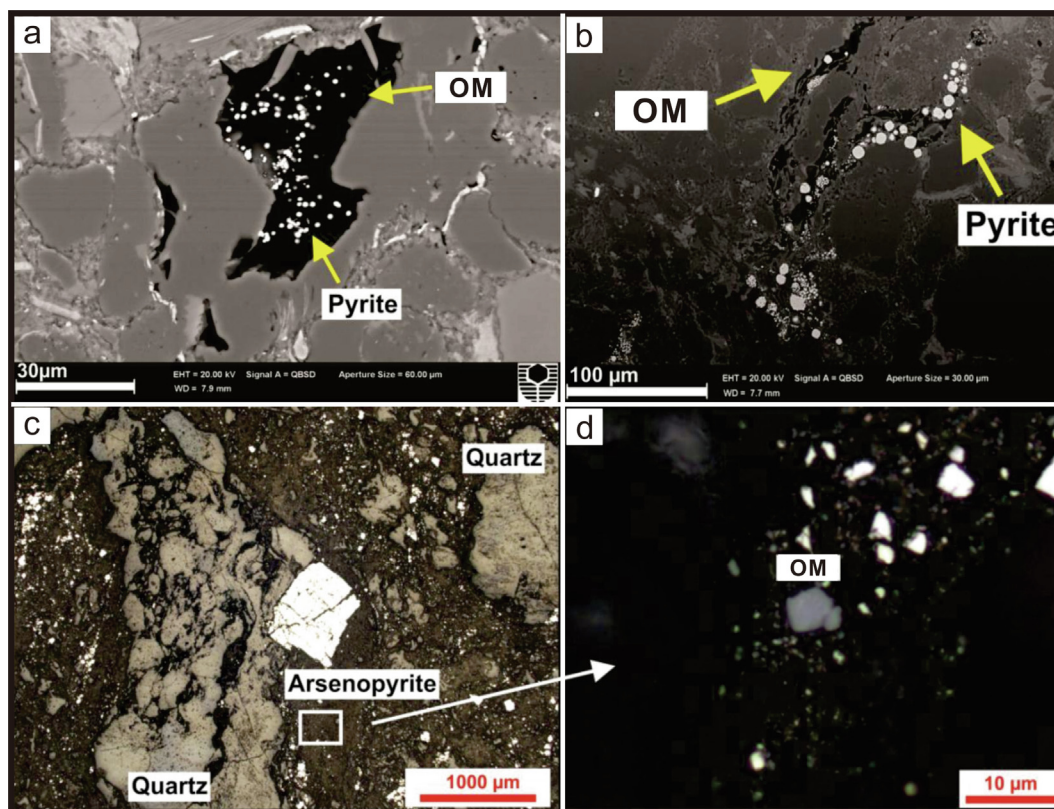
Fig. 15. Relationships among organic matter, sheared rock, and pyrite in metasedimentary orogenic gold deposits, from Hu et al. (2015b): (A) BSE image of organic matter and pyrite; (B) BSE image of strip-shaped organic matter and framboidal pyrite; and (C, D) organic matter particles scattered through the sheared rock with auriferous sulfides.

8. Conclusions

Although usually in nanoscale, NPs are an important part of the Earth surface systems and represent a new hot issue for geological research. The research techniques and origins as well as the properties of NPs in the Earth surface systems are examined, the environment and resource effects of NPs are presented, and the current applications of NPs in environmental remediation and resource exploration are summarized. The main conclusions are as follows.

(1) Techniques for advanced sample treatments and image observations as well as component and isotope analyses have been combined to comprehensively research the morphology, components, structure, origination, and chronology of NPs in the Earth surface systems. Typical NPs including minerals at nanosacle, nano organic matter, natural metal NPs, nanoinclusions, and NPs in soil and water are illustrated. The physical, chemical, microbial, and multigeological origins of NPs are also analyzed and summarized.

(2) NPs have abnormal physicochemical properties including size effect, migration and aggregation effect, adsorption effect, and oxidation–reduction reactivity. Due to these abnormal properties, NPs have environment and resource effects and play an increasingly important role in environmental remediation and concealed deposit prediction. These unique physicochemical properties of NPs may bring about changes in some important geophysical, geochemical, and biogeochemical reactions and kinetic processes during geological processes. Exploring this vast unknown field will expand and enrich our understanding of the geological processes of the Earth surface systems.

(3) Pollutant NPs can bring damage effects to environments such as soil and water. Meanwhile, NPs are applied in the remediation of soil and water pollution by using NPs and modified NPs as well as combined nanomaterial technology. Factors influencing the remediation effects including characters of water and soil, hazardous element species, and NPs themselves are clarified. Future remediation by NPs may be carried out by developing new types of NPs that have larger specific surface areas and greater removal efficiency.

(4) The formation, migration, deposition, and accumulation of NPs are closely related to the resource enrichment and evolution. NPs can be applied to indicate the mineralizing mechanism, reveal concealed deposit information, and help elucidate unconventional reservoir evolution and formation processes. Organic NPs can reflect the generation process of oil and gas, determine the occurrence state of oil and gas, and widely participate in the metallogenic process of metal elements, so as to indicate the coexistence characteristics of organic matter and metal. The influences of the abnormal properties of NPs on the metallogenic processes, formation types, symbiotic relationships, and corresponding metallogenic models of ore deposits need to be further studied.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments:

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