

# Supplementary Information for

Recombinant *Escherichia coli* as a biofactory for various single- and multielement nanomaterials

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Datasets S1 to S2

#### **SI Appendix Materials and Methods**

Bacterial strains and plasmids. All the bacterial strains and plasmids used in this study are listed in SI Appendix, Table S1. Primers used in this study are listed in SI Appendix, Table S3 and were purchased from Macrogen (Seoul, Korea). All the DNA manipulations including restriction enzyme digestion, ligation and agarose gel electrophoresis followed standard procedures (1). For the construction of plasmids and strains, bacteria routinely grew in Luria-Bertani (LB) broth containing (per liter) 10 g tryptone, 5 g yeast extract and 10 g NaCl or on LB-agar medium containing 1.5% (w/v) agar. Kanamycin (Km; 30 μg ml<sup>-1</sup>) was added to the medium when required. Polymerase chain reactions (PCRs) were conducted using C1000 Touch<sup>TM</sup> Thermal Cycler (Bio-Rad, Hercules, CA, USA). Restriction enzymes were purchased from New England Biolabs (Ipswich, MA, USA) or Enzynomics (Daejeon, Korea). To clone the MT gene, the DNA fragment encoding bacterial MT corresponding to PP\_3262 gene of Pseudomonas putida KT2440 was amplified by PCR using primers P1 and P2 with the genomic DNA of P. putida KT2440 as a template. The amplified MT gene fragment was digested using EcoRI and PstI and was ligated to plasmid pTac15K digested using the same restriction enzymes, resulting in plasmid pYJ-MT (SI Appendix, Table S1). To clone the PCS gene, the DNA fragment encoding PCS in Arabidopsis thaliana ecotype Columbia (leaves) was amplified by PCR using primers P3 and P4 with the cDNA library of A. thaliana as a template. The amplified PCS gene was digested using PstI and SalI and was ligated to plasmid pTac15K digested using the same restriction enzymes, constructing pYJ-PCS (SI Appendix, Table S1). To coexpress MT and PCS genes in a single plasmid, a gene fragment containing tac promoter, MT gene and rrnB terminator in the plasmid pYJ-MT as a template and tac promoter and *PCS* were ligated using the primers P5 and P6, constructing a co-expression plasmid pYJ-MT-PCS (*SI Appendix*, Table S1). Colonies were selected on LB-agar plates with Km supplemented. Recombinant strains harboring correct plasmid pYJ-MT-PCS were selected by colony PCR and were further confirmed by sequencing.

Induction of MT and PCS expression. Seed culture of the recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS was prepared in 25-ml test tubes containing 10 ml LB medium supplemented with Km at 37 °C overnight in a rotary shaker with continuous shaking at 200 rpm. One milliliter of overnight culture was transferred to 100 ml of LB medium supplemented with Km in a 250-ml Erlenmeyer flask. Cell growth was monitored by measuring the absorbance at 600 nm (OD<sub>600</sub>) using Ultrospec 3000 spectrophotometer (Amersham Biosciences, Uppsala, Sweden). When OD<sub>600</sub> reached about 0.6, the co-expression of MT and PCS in the cells was induced by the adding 0.5 mM of isopropyl  $\beta$ -D-1-thiogalactopyranoside (IPTG; Sigma-Aldrich, St. Louis, MO, USA). The induced cells were further incubated depending on purposes.

**Expression of MT and PCS.** The expression of MT and PCS was examined with tricinesodium dodecyl sulfate polyacrylamide gel electrophoresis (SDS-PAGE) (Schägger, 2006). To prepare samples for tricine-SDS-PAGE, *E. coli* DH5 $\alpha$  and *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS were incubated in 25-ml tubes containing10 ml of LB medium and Km at 37°C and 200 rpm for 12 h. After seed culture, cells were transferred to a 250 ml flask containing 100 ml of LB medium. When the OD<sub>600</sub> was reached at 0.6, 0.5 mM, IPTG was added for induction of MT and PCS. After induction of MT and PCs expression, 0.5 mM of Sn precursor (i.e.  $SnCl_2 \cdot 2H_2O$ ) was added and further incubated for 12 h. Harvested cell pellets were resuspended in 300 µL of lysis buffer (20 mM Tris-HCl pH 8.0, 300 mM NaCl and 5 mM imidazole) in 1.5 ml microcentrifuge tubes to give a final  $OD_{600}$  of 1. The resuspended cells were lysed by sonication and boiled at 100°C for 5 min after addition of the loading buffer. The samples were electrophoresed on 16% tricine-SDS-PAGE gel and were analyzed after staining with Coomassie Brilliant Blue R-250 for 60 min at 80 V and for 30 min at 250 V.

*In vivo* synthesis of NMs. For *in vivo* synthesis of NM, the recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS induced for MT and PCS co-expression was further cultivated for 1 h and then corresponding precursors (*SI Appendix*, Table S4) were supplemented to the medium at final concentrations of 0.5 mM. After supplementing the precursors, the pH of the culture medium range from 6.3 to 6.5. To exclude any possible impact of different initial pH on NM biosynthesis while minimizing the possible influence on the NM synthesis during pH adjusting procedure with OH<sup>-</sup> ions, the pH of the culture medium was standardized to pH 6.5 using 1 M NaOH solution, which requires minimum amount of base. After additional cultivation at 37 °C in a rotary shaker with continuous shaking at 200 rpm for 12 h. The culture medium was centrifuged at 1,290*g* for 15 min at 4 °C to collect the NMs synthesized from the recombinant cells. The supernatant was discarded and the pellet was resuspended in distilled water.

**Cell growth of recombinant** *E. coli* **in the presence of different precursors**. Seed culture of the recombinant *E. coli* DH5α harboring pYJ-MT-PCS was prepared by growing cells

in 25 mL test tubes containing 10 mL of LB medium supplemented with Km at 37°C and 200 rpm overnight in a rotary shaker. One mL of overnight culture was transferred to 250 mL flask containing100 mL of LB medium supplemented with Km. Cell growth was monitored by measuring the OD<sub>600</sub> using UV-vis spectrophotometer. When OD<sub>600</sub> reached about 0.6, cells were induced with 0.5 mM of IPTG for MT and PCS expression and further cultivated for 1 h. Next, the corresponding precursors (i.e. Ag, Sn, Fe, Ni, Ag/S and Fe/Ni) were supplemented to the medium at a final concentration of 0.5 mM. Cells were cultured at 37°C and 200 rpm for additional 12 h (Fig. S1).

**Preparation of cell extract.** Recombinant *E. coli* cells were harvested after cultivation by centrifugation at 1,290g for 15 min at 4 °C and washed three times with 10 ml each of wash buffer containing (per liter) 181.8 11.25 g g urea. glycine, 0.37 g ethylenediaminetetraacetic acid and 0.77 g dithiothreitol. The washed pellet was resuspended in distilled water and disrupted by sonication using Vibra-Cell VCX 600 (Sonics & Materials Inc., Newtown, CT, USA) equipped with a titanium probe (13 mm, Sonics and Materials Inc., Newtown, CT, USA). Cell debris was separated and removed by centrifugation at 1,290g for 15 min at 4 °C. The soluble proteins in the final cell extract solutions were quantified by the Bio-Rad Protein Assay Kit (Bio-Rad, Hercules, CA, USA) with bovine serum albumin as a standard.

*In vitro* synthesis of NMs. To prepare cell extract for *in vitro* NM biosynthesis, the recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS induced for MT and PCS co-expression was further cultivated for 12 h. Cell extract was prepared from the resulting cells. For the

*in vitro* NM synthesis, various precursors for NM biosynthesis (*SI Appendix*, Table S4) were added at a final concentration of 1 mM into the cell extract diluted with distilled water (protein concentration of 1 mM) in a 50-ml test tubes. The pH of *in vitro* reaction solution was adjusted to 6.5 as same to *in vivo* NM synthesis, using 1 M NaOH solution. The precursors and the cell extract were incubated at 25 °C in a rotary shaker for 12 h with shaking at 200 rpm.

*In vivo* synthesis of crystalline NMs at initial pH 7.5. To synthesize crystalline NMs *in vivo* at higher initial pH of 7.5, the recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS induced for MT and PCS co-expression were further cultivated for 1 h with IPTG induction and corresponding precursor (*SI Appendix*, Table S4) was added to a final concentration of 0.5 mM into the culture medium. The pH of culture medium was adjusted to pH 7.5 using 1M NaOH solution. After further cultivation for 12 h, the culture medium was centrifuged at 1,290g and 4 °C for 15 min to collect the NMs from the recombinant cells. The supernatant was discarded and the pellet was resuspended in distilled water.

*In vitro* synthesis of crystalline NM at initial pH 7.5. To synthesize crystalline NM *in vitro* at increase initial pH, the recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS induced for MT and PCS co-expression was further cultivated for 12 h. Cell extract was prepared from the resulting cells. Corresponding precursor (*SI Appendix*, Table S4) was added at final concentrations of 1 mM into the cell extract diluted with distilled water to its protein concentration of 1 mM in a 50-ml test tubes. The pH of *in vitro* reaction solution was

adjusted to pH 7.5 using 1 M NaOH solution. The precursors and the cell extract were incubated at 25 °C in a rotary shaker for 12 h with shaking at 200 rpm.

In vivo and in vitro synthesis of crystalline a-Fe<sub>2</sub>O<sub>3</sub> NM. To biosynthesize crystalline a- $Fe_2O_3$  NM *in vivo* the recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS induced for MT and PCS co-expression were further cultivated for 1 h with IPTG induction and the Fe precursor (FeCl<sub>2</sub>·6H<sub>2</sub>O) was added to the culture medium (final ion concentration of 0.5 mM). The pH of culture medium was adjusted to pH 6.5 using 1M NaOH solution. After further cultivation at 37 °C in a rotary shaker for 6 h with shaking at 200 rpm, the culture medium was centrifuged at 1,290g and 4 °C for 15 min to collect the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>,  $\beta$ -FeOOH and  $\alpha$ -FeOOH NMs from the recombinant cells. The supernatant was discarded and the pellet was resuspended in distilled water. To obtain  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM, the two byproducts of  $\beta$ -FeOOH and α-FeOOH were removed by calcination at 800 °C for 2 h in the presence of air with a heating rate of  $2^{\circ}$  min<sup>-1</sup>. To obtain crystalline  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM *in vitro*, the recombinant *E. coli* DH5α harboring pYJ-MT-PCS induced for MT and PCS co-expression was further cultivated at 37 °C in a rotary shaker for 12 h with shaking at 200 rpm. Cell extract was prepared from the resulting cells. The FeCl<sub>2</sub>·6H<sub>2</sub>O was added to the final ion concentration of 1 mM into the cell extract diluted with distilled water (protein concentration of 1 mM) in a 50-ml test tubes. The pH of in vitro reaction solution was set at pH 6.5 using 1M NaOH solution. The precursor and the cell extract were incubated at 25 °C in a rotary shaker at 200 rpm for 6 h. To obtain α-Fe<sub>2</sub>O<sub>3</sub> NM, calcination was performed as before.

Characterization of the biosynthesized NMs. The structure and morphology of the biosynthesized NMs were observed by field emission transmission electron microscopy (TEM); Tecnai F20 and Tecnai F30 (FEI Company, Frankfurt, Germany) were operated at accelerating voltages of 200 and 300 kV, respectively. The biosynthesized NMs for TEM samples were prepared by dispersing them in distilled water on a carbon-Cu grid, except for the biosynthesized Cu NM which was placed on a Ni grid. The average sizes of the biosynthesized NMs were determined by TEM analysis. Each grain up to a certain maximum diameter was manually measured to minimize counting errors one-by-one and sequentially recorded for about 100 NMs. Measurements of interplanar distance of biosynthesized crystalline NMs lattice were performed using DiffTools suite in DigitalMicrograph software (3). To identify element composition of the biosynthesized NMs, energy dispersive spectrometer (EDS) analysis was performed simultaneously. For X-ray diffraction (XRD) analysis, the NMs synthesized in vivo were resuspended in distilled water and disrupted by sonication. Then, they were centrifuged at  $16,100 \times g$  and 4 °C for 10 min to collect the precipitates. The supernatant was discarded, and the precipitates were then resuspended in distilled water. In the case of NMs synthesized in *vitro*, the samples were resuspended in distilled water and subsequently centrifuged at 16,100  $\times g$  and 4 °C for 10 min to collect the precipitates. The supernatant was discarded, and the precipitates were resuspended in distilled water. The prepared samples were dropcast on a glass substrate and dried in 70 °C oven. We repeated these steps for several times until a thick film or powder was obtained. The XRD patterns were characterized by X-ray diffractometer (D/MAX-2,500, Rigaku, Japan) with CuKa radiation ( $\lambda$ =1.5406Å), 2 $\Theta$ range 20-70° and a scanning rate of 2° min<sup>-1</sup>. Surface characterization of biosynthesized NMs was conducted in the form of KBr tablets by Fourier transform infrared (FTIR) spectrometry (Nicolet<sup>TM</sup> iS<sup>TM</sup>50, Thermo Scientific, Waltham, MA, USA). The magnetization of biosynthesized magnetic crystalline NMs, including Mn<sub>3</sub>O<sub>4</sub>, Fe<sub>3</sub>O<sub>4</sub>, CoFe<sub>2</sub>O<sub>4</sub>, NiFe<sub>2</sub>O<sub>4</sub>, ZnMn<sub>2</sub>O<sub>4</sub> and ZnFe<sub>2</sub>O<sub>4</sub> were investigated by superconducting quantum interference device-vibrating sample magnetometer (SQUID-VSM; Quantum Design, San Diego, CA, USA). The magnetic properties were measured at 300 K in an applied magnetic field sweeping from -50,000 Oe to +50,000 Oe. To remove surface-bound functional groups, *in vivo* synthesized magnetic crystalline NMs were calcinated at 800 °C for 2 h in the presence of air with a heating rate of 2° min<sup>-1</sup>.

**Generation of Pourbaix diagrams.** To predict NM producibility, Pourbaix diagram analyses of each element at an ionic concentration of 0.5 (*in vivo*) or 1 mM (*in vitro*) were performed in aqueous solution at 37 °C (*in vivo*) or 25 °C (*in vitro*) with the Eh range of -1.046 to +1.046 V and pH range of 6 to 10 using Hydra/Medusa chemical equilibrium database and plotting software (4). It has been reported that precursor ions can form complexes and thus interfere with NM biosynthesis (5, 6). The input for generating Pourbaix diagrams includes all the precursor species and their concentrations, and temperatures and pressure (1 bar) of each *in vivo* or *in vitro* biosynthesis condition in the database. The results of Pourbaix diagrams can vary depending on the conditions such as the precursor species and their concentrations, temperatures, and pressures, respectively.

**Measurement of Eh and pH.** The Eh and pH were measured during *in vivo* and *in vitro* biosynthesis of NM of each element for 12 h. The time series measurements (0, 1, 2, 4, 6,

9 and 12 h) of the Eh and pH values were performed using electrochemistry benchtop meters (Orion <sup>TM</sup> star A211, Thermo Scientific,S11 Waltham, MA, USA).

**Measurement of NADH level.** Seed cultures of the wildtype *E. coli* DH5 $\alpha$  strain harboring pTac15K (control) and recombinant *E. coli* DH5 $\alpha$  strain harboring pYJ-MT-PCS were prepared in 25-ml test tubes containing 10 ml LB medium and 30 µg ml<sup>-1</sup> of Km (for recombinant strain) at 37 °C overnight in a rotary shaker at 200 rpm. One milliliter of overnight culture was used to inoculate a 250-ml flask containing 100 ml of LB medium; for recombinant strain, Km (30 µg ml<sup>-1</sup>) was supplemented. Cells were grown to an OD<sub>600</sub> of 0.6, and were induced with 0.5 mM of IPTG. Cells were further cultivated for 1 h after IPTG induction. The NADH levels in the wildtype and recombinant strains were measured at 0 and 12 h using a colorimetric kit (K337-100, BioVision, Milpitas, CA, USA) by recording the absorbance at 450 nm (SpectraMax M2, Molecular Devices, Sunnyvale, CA, USA). The NADH levels of cell extract were measured using the cell extract prepared as described above. Measurements of NADH levels were conducted in triplicates.

## **SI Appendix Figures**



**Fig. S1.** Growth curves of *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS cells in the presence of 0.5 mM of Ag, Sn, Fe, Ni, Ag/S and Fe/Ni. The control (*E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS) growth curve without precursor is shown with black circles. As can be seen, the extent of growth inhibition varies with different ions.



**Fig. S2.** TEM images and EDS graphs of the nine crystalline NMs synthesized *in vivo*. (*A*), TEM images of the crystalline NMs synthesized *in vivo* with their corresponding elements labeled in green circle. Scale bar, 20 nm. (*B*), The EDS graphs of the crystalline NMs synthesized *in vivo* with their corresponding elements labeled in green square. The presence of P signal is likely due to the presence of cell debris bound to the surface of the crystalline NMs. There was Cu NM placed on a Ni grid. "\*\*" indicates the presence of minor peaks, at around 6.4 keV and 6.9 keV from the instrument.



**Fig. S3.** HR-TEM images and the XRD patterns of the nine crystalline NMs synthesized *in vivo*. (*A*) HR-TEM images of the crystalline NMs synthesized *in vivo* with their corresponding elements labeled in green circle. The interplanar distance of the biosynthesized crystalline NM lattice and the Miller indices of crystallographic planes within parentheses are shown together. (*B*) The crystalline compositions of the biosynthesized crystalline NMs were analyzed by XRD. The green numbers in parenthesis represent the joint committee on powder diffraction standards (JCPDS) cards. The black numbers in parentheses represent Miller indices corresponding to each scattering peak. Because cell debris existed for NMs synthesized *in vivo*, background scattering from the some organic components was also recorded in the XRD patterns.



**Fig. S4.** Reconstructed crystal structures of the eight biosynthesized crystalline NMs. Illustrations of crystalline structures of the biosynthesized crystalline NMs based on the XRD data. Colored spheres and sticks represent individual atoms and chemical bonds between the atoms, respectively.



**Fig. S5.** TEM images and EDS graphs of the 12 amorphous NMs synthesized *in vivo*. (*A*) TEM images of the amorphous NMs synthesized *in vivo* with their corresponding elements labeled in yellow circle. Scale bar, 20 nm. (*B*) The EDS graphs of the amorphous NMs synthesized *in vivo*. The amorphous NMs synthesized *in vivo* with their corresponding elements labeled in yellow square.



**Fig. S6.** SDS-PAGE analysis of MT and PCS expression in *E. coli* DH5 $\alpha$  strain. Expression of MT (~7.9 kDa) and PCS (~54.5 kDa) in *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS strain during cultivation. Red arrow indicates the PCS band (~54.5 kDa). The lanes are: L, molecular weight marker (kDa); S1, soluble protein fraction of *E. coli* DH5 $\alpha$  cultivated for 12 h; S2, soluble protein fraction of recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS cultivated for 2 h (before induction); S3-S6, soluble protein fractions of the recombinant *E. coli* at 2, 4, 8 and 12 h post induction in the presence of Sn precursor, respectively.



**Fig. S7.** TEM images and EDS graphs of the seven crystalline NMs synthesized *in vitro*. (*A*) TEM images of the crystalline NMs synthesized *in vitro* with their corresponding elements labeled in purple circle. Scale bar, 20 nm. (*B*) The EDS graphs of the crystalline NMs synthesized *in vitro*. The amorphous NMs synthesized *in vitro* with their corresponding elements labeled in purple square.



**Fig. S8.** HR-TEM images and the XRD patterns of the seven crystalline NMs synthesized *in vitro*. (*A*) HR-TEM images of the crystalline NMs synthesized *in vitro* with their corresponding elements labeled in purple circle. The interplanar distance of the crystalline NM lattice and the Miller indices of crystallographic planes within parentheses are shown together. (*B*) The crystalline composition confirmation of the crystalline NMs synthesized *in vitro* were analyzed using XRD. The purple numbers in parentheses represent the JCPDS cards. The black numbers in parentheses represent Miller indices corresponding to each scattering peak.



**Fig. S9.** TEM images and EDS graphs of the four amorphous NMs synthesized *in vitro*. (*A*) TEM images of the amorphous NMs synthesized *in vitro* with their corresponding elements labeled in pink circle. Scale bar, 20 nm. (*B*) The EDS graphs of the amorphous NMs synthesized *in vitro*. The amorphous NMs synthesized *in vitro* with their corresponding elements labeled in pink square.



**Fig. S10.** NADH levels in wildtype *E. coli* DH5 $\alpha$  strain harboring pTac15K, recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS and that of their cell extract. NADH levels *in vivo* were measured in wildtype *E. coli* DH5 $\alpha$  harboring a control vector pTac15K and the recombinant *E. coli* DH5 $\alpha$  harboring pYJ-MT-PCS under 37°C at 0 h and 12 h. The NADH levels *in vitro* were measured from the cell extracts of the above strains under 25°C at 0 h and 12 h. Experiments were conducted in triplicates. Error bar indicates standard deviation (s.d).



**Fig. S11.** Pourbaix diagram analyses and measurements of the Eh and pH during *in vivo* biosynthesis of nine crystalline NMs. In Pourbaix diagram analyses, the upper blue dashed line denotes the Eh for the oxygen evolution reaction, while the lower blue dashed line corresponds to the Eh for the hydrogen evolution reaction. Between the two blue dashed lines represent the stability region of H<sub>2</sub>O. The phase type is indicated at the end of the name, such as (c) or (cr) for crystalline and (s) for solid. The chemical species taken into account include: MnO<sub>4</sub><sup>-</sup>, MnO<sub>2</sub> (cr), Mn<sub>2</sub>O<sub>3</sub> (cr), Mn<sub>3</sub>O<sub>4</sub> (c), Mn<sup>2+</sup>, Mn<sub>2</sub>(OH)<sub>3</sub><sup>+</sup> and Mn(OH)<sub>2</sub> for Mn; Fe<sub>2</sub>O<sub>3</sub> (c), Fe<sub>3</sub>O<sub>4</sub> (c), Fe<sup>2+</sup>, Fe(OH)<sub>2</sub> (c) and Fe (c) for Fe; CuO (c), Cu<sub>2</sub>O (c) and Cu (c) for Cu; MoO<sub>4</sub><sup>2-</sup>, MoO<sub>2</sub> (c) and Mo (c) for Mo; Ag<sub>2</sub>O<sub>3</sub> (c), AgO (c), Ag<sup>+</sup>, Ag<sub>2</sub>O (c) and Ag (c) for Ag; In(OH)<sub>3</sub> (c) and In (s) for In; SnO<sub>2</sub> (c) and Sn (s) for Sn; H<sub>2</sub>TeO<sub>4</sub>, HTeO<sub>4</sub><sup>-</sup>, TeO(OH)<sub>3</sub><sup>-</sup>, TeO<sub>2</sub>(OH)<sub>2</sub><sup>2-</sup>, Te (c) and Te<sub>2</sub><sup>2-</sup> for Te; Au(OH)<sub>3</sub> (c), AuOH and Au (c) for Au. The Eh and pH of the *in vivo* reactions were mapped (shown in the circle with the pink to red). The compositions of the *in vivo* synthesized crystalline NMs according to the XRD results [Mn<sub>3</sub>O<sub>4</sub>, Fe<sub>3</sub>O<sub>4</sub>, Cu<sub>2</sub>O, Ag, In(OH)<sub>3</sub>, SnO<sub>2</sub>, Te and Au], which are indicated by the yellow box.



**Fig. S12.** Pourbaix diagram analyses and measurements of the Eh and pH during *in vivo* biosynthesis of 12 amorphous NMs. The chemical species taken into account include:  $SeO_4^{2^-}$ ,  $HSeO_3^{2^-}$ ,  $SeO_3^{2^-}$ , Se (c) and  $HSe^-$  for Se;  $Y^{3^+}$  and  $Y(OH)_3$  (c) for Y;  $ZrO_2$  (c) for Zr;  $La^{3^+}$  and  $La(OH)_3$  (c) for La;  $CeO_2$  (c),  $Ce^{3^+}$  and  $Ce(OH)_3$  (c) for Ce;  $Pr^{3^+}$  and  $Pr(OH)_3$  (c) for Pr;  $Nd^{3^+}$  and  $Nd(OH)_3$  (c) for Nd;  $Sm^{3^+}$  and  $Sm(OH)_3$  (c) for Sm;  $Eu^{3^+}$ ,  $Eu(OH)_3$  (c) and  $Eu^{2^+}$  for Eu;  $Gd^{3^+}$  and  $Gd(OH)_3$  (c) for Gd; HgO (c) and Hg (l) for Hg; PbO<sub>2</sub> (c), Pb(OH)<sub>2</sub> (c) and Pb (c) for Pb. The Eh and pH of the *in vivo* reactions were mapped (shown in the circle with the pink to red).



**Fig. S13.** Pourbaix diagram analyses and measurements of the Eh and pH in the *in vivo* conditions of no NM formation. The chemical species taken into account include: Li<sup>+</sup> for Li; B(OH)<sub>3</sub> and H<sub>2</sub>BO<sub>3</sub> for B; Al(OH)<sub>3</sub> (cr) for Al; VO<sub>2</sub>(OH)<sub>2</sub><sup>-</sup>, VO<sub>3</sub>OH<sup>2-</sup>, V<sub>6</sub>O<sub>13</sub> (c), V<sub>2</sub>O<sub>4</sub> (c), V<sub>3</sub>O<sub>5</sub> (c), V<sub>2</sub>O<sub>3</sub> (c) and VOH<sup>+</sup> for V; HCrO<sub>4</sub><sup>-</sup>, CrO<sub>2</sub> (c), CrO<sub>4</sub><sup>2-</sup>, Cr<sub>2</sub>O<sub>3</sub> (cr) and Cr<sup>2+</sup> for Cr; Co(OH)<sub>3</sub> (c), Co<sup>2+</sup>, Co(OH)<sub>2</sub> (c) and Co (c) for Co; NiO<sub>2</sub> (s), Ni<sup>2+</sup>, Ni(OH)<sub>2</sub> (c) and Ni (c) for Ni; Zn<sup>2+</sup>, ZnO (cr) and Zn (c) for Zn; Rb<sup>+</sup> for Rb; Sr<sup>2+</sup> for Sr; Y<sup>3+</sup> and Y(OH)<sub>3</sub> (c) for Y; Cd<sup>2+</sup>, Cd(OH)<sub>2</sub> (cr) and Cd (c) for Cd; Ba<sup>2+</sup> and BaCO<sub>3</sub> (c) for Ba; HW<sub>6</sub>O<sub>21</sub><sup>5-</sup>, WO<sub>4</sub><sup>2-</sup>, WO<sub>2</sub> (c) and W (c) for W. The Eh and pH of the *in vivo* reactions were mapped (shown in the circle with the pink to red).



**Fig. S14.** Pourbaix diagram analyses and measurements of the Eh and pH during *in vitro* biosynthesis of seven crystalline NMs. The chemical species taken into account include:  $MnO_4^-$ ,  $MnO_2$  (cr),  $Mn_2O_3$  (cr),  $Mn_3O_4$  (c),  $Mn^{2+}$ ,  $Mn_2(OH)_3^+$  and  $Mn(OH)_2$  for Mn; Fe<sub>2</sub>O<sub>3</sub> (c), Fe<sub>3</sub>O<sub>4</sub> (c), Fe<sup>2+</sup>, Fe(OH)<sub>2</sub> (c) and Fe (c) for Fe; CuO (c), Cu<sub>2</sub>O (c) and Cu (c) for Cu;  $MoO_4^{2-}$ ,  $MoO_2$  (c) and Mo (c) for Mo; Ag<sub>2</sub>O<sub>3</sub> (c), AgO (c), Ag<sup>+</sup>, Ag<sub>2</sub>O (c) and Ag (c) for Ag; In(OH)<sub>3</sub> (c) and In (s) for In; SnO<sub>2</sub> (c) and Sn (s) for Sn; Au(OH)<sub>3</sub> (c), AuOH and Au (c) for Au. The Eh and pH of the *in vitro* reactions were mapped (shown in the circle with the pink to red). The compositions of the *in vitro* synthesized crystalline NMs according to the XRD results [Mn<sub>3</sub>O<sub>4</sub>, Fe<sub>3</sub>O<sub>4</sub>, Cu<sub>2</sub>O, Ag, In(OH)<sub>3</sub>, SnO<sub>2</sub> and Au], which are indicated by the yellow box.



**Fig. S15.** Pourbaix diagram analyses and measurements of the Eh and pH during *in vitro* biosynthesis of four amorphous NMs. The chemical species taken into account include:  $SeO_4^{2^-}$ ,  $HSeO_3^{2^-}$ ,  $SeO_3^{2^-}$ , Se(c) and  $HSe^-$  for Se;  $ZrO_2$  (c) for Zr;  $H_2TeO_4$ ,  $HTeO_4^-$ ,  $TeO(OH)_3^-$ ,  $TeO_2(OH)_2^{2^-}$ , Te(c) and  $Te_2^{2^-}$  for Te;  $PbO_2(c)$ ,  $Pb(OH)_2(c)$  and Pb(c) for Pb. The Eh and pH of the *in vitro* reactions were mapped (shown in the circle with the pink to red).



**Fig. S16.** Pourbaix diagram analyses and measurements of the Eh and pH in the *in vitro* conditions of no NM formation. The chemical species taken into account include: Li<sup>+</sup> for Li; B(OH)<sub>3</sub> and H<sub>2</sub>BO<sub>3</sub> for B; Al(OH)<sub>3</sub> (cr) for Al; VO<sub>2</sub>(OH)<sub>2</sub><sup>-</sup>, VO<sub>3</sub>OH<sup>2-</sup>, V<sub>6</sub>O<sub>13</sub> (c), V<sub>2</sub>O<sub>4</sub> (c), V<sub>3</sub>O<sub>5</sub> (c), V<sub>2</sub>O<sub>3</sub> (c) and VOH<sup>+</sup> for V; HCrO<sub>4</sub><sup>-</sup>, CrO<sub>2</sub> (c), CrO<sub>4</sub><sup>2-</sup>, Cr<sub>2</sub>O<sub>3</sub> (cr) and Cr<sup>2+</sup> for Cr; Co(OH)<sub>3</sub> (c), Co<sup>2+</sup>, Co(OH)<sub>2</sub> (c) and Co (c) for Co; NiO<sub>2</sub> (s), Ni<sup>2+</sup>, Ni(OH)<sub>2</sub> (c) and Ni (c) for Ni; Zn<sup>2+</sup>, ZnO (cr) and Zn (c) for Zn; Rb<sup>+</sup> for Rb; Sr<sup>2+</sup> for Sr; Y<sup>3+</sup> and Y(OH)<sub>3</sub> (c) for Y; MoO<sub>4</sub><sup>2-</sup>, MoO<sub>2</sub> (cr) and Mo (c) for Mo; Cd<sup>2+</sup>, Cd(OH)<sub>2</sub> (c) and Cd (c) for Cd; Ba<sup>2+</sup> and BaCO<sub>3</sub> (c) for Ba; La<sup>3+</sup> and La(OH)<sub>3</sub> (c) for La; CeO<sub>2</sub> (c), Ce<sup>3+</sup> and Ce(OH)<sub>3</sub> (c) for Ce; Pr<sup>3+</sup> and Pr(OH)<sub>3</sub> (c) for Fr; Nd<sup>3+</sup> and Nd(OH)<sub>3</sub> (c) for Nd; Sm<sup>3+</sup> and Sm(OH)<sub>3</sub> (c) for Sm; Eu<sup>3+</sup>, Eu(OH)<sub>3</sub> (c) and Eu<sup>2+</sup> for Eu; Gd<sup>3+</sup> and Gd(OH)<sub>3</sub> (c) for Gd; HW<sub>6</sub>O<sub>21</sub><sup>5-</sup>, WO4<sup>2-</sup>, WO<sub>2</sub> (c) and W (c) for W; HgO (c) and Hg (l) for Hg. The Eh and pH of the *in vitro* reactions were mapped (shown in the circle colored in pink to red).



**Fig. S17.** Pourbaix diagram analyses and measurements of the Eh and pH during *in vivo* biosynthesis of 12 crystalline NMs at pH 7.5. The chemical species taken into account include: Co(OH)<sub>3</sub> (c), Co<sup>2+</sup>, Co(OH)<sub>2</sub> (c) and Co (c) for Co; NiO<sub>2</sub> (s), Ni<sup>2+</sup>, Ni(OH)<sub>2</sub> (c) and Ni (c) for Ni; Zn<sup>2+</sup>, ZnO (cr) and Zn (c) for Zn; Cd<sup>2+</sup>, Cd(OH)<sub>2</sub> (c) and Cd (c) for Cd; Ba<sup>2+</sup> and BaCO<sub>3</sub> (c) for Ba; La<sup>3+</sup> and La(OH)<sub>3</sub> (c) for La; CeO<sub>2</sub> (c), Ce<sup>3+</sup> and Ce(OH)<sub>3</sub> (c) for Ce; Pr<sup>3+</sup> and Pr(OH)<sub>3</sub> (c) for Pr; Nd<sup>3+</sup> and Nd(OH)<sub>3</sub> (c) for Nd; Sm<sup>3+</sup> and Sm(OH)<sub>3</sub> (c) for Sm; Eu<sup>3+</sup>, Eu(OH)<sub>3</sub> (c) and Eu<sup>2+</sup> for Eu; Gd<sup>3+</sup> and Gd(OH)<sub>3</sub> (c) for Gd. The Eh and pH of the *in vivo* reactions were mapped (shown in the circle with the pink to red). The compositions of *in vivo* synthesized crystalline NMs at pH 7.5 according to the XRD results [β-Co(OH)<sub>2</sub> and Co<sub>3</sub>O<sub>4</sub>, β-Ni(OH)<sub>2</sub>, ZnO, β-Cd(OH)<sub>2</sub>, Cd(OH)<sub>2</sub>, BaCO<sub>3</sub>, La(OH)<sub>3</sub>, CeO<sub>2</sub> and Ce(OH)<sub>3</sub>, Pr(OH)<sub>3</sub>, Nd(OH)<sub>3</sub>, Sm(OH)<sub>3</sub>, Eu(OH)<sub>3</sub> and Gd(OH)<sub>3</sub>], which are indicated by the yellow box.



**Fig. S18.** TEM images and EDS graphs of the 12 crystalline NMs synthesized *in vivo* at the pH 7.5. (*A*) TEM images of the crystalline NMs synthesized *in vivo* with their corresponding elements labeled in red circle. Scale bar, 20 nm. (*B*) The EDS graphs of the crystalline NMs synthesized *in vivo*. The crystalline NMs synthesized *in vivo* with their corresponding elements labeled in red square.



**Fig. S19.** HR-TEM images and the XRD patterns of 12 crystalline NMs synthesized *in vivo* at pH 7.5. (*A*) HR-TEM images of the crystalline NMs synthesized *in vivo* at pH 7.5 with their corresponding elements labeled in red circle. The interplanar distance of the crystalline NM lattice and the Miller indices of crystallographic planes within parentheses are shown together. (*B*) The crystalline compositions of the biosynthesized crystalline NMs were analyzed using XRD. The red numbers in parentheses represent JCPDS cards. The black numbers in parentheses represent Miller indices corresponding to each scattering peak. Because cell debris existed for NMs synthesized *in vivo*, background scattering from the some organic components was also recorded in the XRD patterns.



**Fig. S20.** Pourbaix diagram analyses and measurements of the Eh and pH in the *in vivo* conditions of amorphous NMs or no NM formation at pH 7.5. (*A*) Formation of amorphous NMs at pH at 7.5. The species taken into account include:  $SeO_4^{2-}$ ,  $HSeO_3^{2-}$ ,  $SeO_3^{2-}$ , Se (c) and HSe<sup>-</sup> for Se; Y<sup>3+</sup> and Y(OH)<sub>3</sub> (c) for Y; ZrO<sub>2</sub>(c) for Zr; HgO (c) and Hg (l) for Hg; PbO<sub>2</sub> (c), Pb(OH)<sub>2</sub> (c) and Pb (c) for Pb. (*B*) No NM synthesis at pH 7.5. The species taken into account include: B(OH)<sub>3</sub> and H<sub>2</sub>BO<sub>3</sub> for B; Al(OH)<sub>3</sub> (cr) for Al; VO<sub>2</sub>(OH)<sub>2</sub><sup>-</sup>, VO<sub>3</sub>OH<sup>2-</sup>, V<sub>6</sub>O<sub>13</sub> (c), V<sub>2</sub>O<sub>4</sub> (c), V<sub>3</sub>O<sub>5</sub> (c), V<sub>2</sub>O<sub>3</sub> (c) and VOH<sup>+</sup> for V; HCrO<sub>4</sub><sup>-</sup>, CrO<sub>2</sub> (c), CrO<sub>4</sub><sup>2-</sup>, Cr<sub>2</sub>O<sub>3</sub> (cr) and Cr<sup>2+</sup> for Cr. The Eh and pH of the *in vivo* reactions were mapped (shown in the circle with the pink to red).



**Fig. S21.** TEM images and EDS graphs of the amorphous NMs synthesized *in vivo* at pH 7.5. (*A*) TEM images of the amorphous NMs synthesized *in vivo* with their corresponding elements labeled in black circle. Scale bar, 20 nm. (*B*) The EDS graphs of the amorphous NMs synthesized *in vivo*. The amorphous NMs synthesized *in vivo* with their corresponding elements labeled in black square.



**Fig. S22.** Pourbaix diagram analyses and measurements of the Eh and pH during *in vitro* biosynthesis of 12 crystalline NMs at pH 7.5. The chemical species taken into account include: Co(OH)<sub>3</sub> (c), Co<sup>2+</sup>, Co(OH)<sub>2</sub> (c) and Co (c) for Co; NiO<sub>2</sub> (s), Ni<sup>2+</sup>, Ni(OH)<sub>2</sub> (c) and Ni (c) for Ni; Zn<sup>2+</sup>, ZnO (cr) and Zn (c) for Zn; Cd<sup>2+</sup>, Cd(OH)<sub>2</sub> (c) and Cd (c) for Cd; La<sup>3+</sup> and La(OH)<sub>3</sub> (c) for La; CeO<sub>2</sub> (c), Ce<sup>3+</sup> and Ce(OH)<sub>3</sub> (c) for Ce; Pr<sup>3+</sup> and Pr(OH)<sub>3</sub> (c) for Pr; Nd<sup>3+</sup> and Nd(OH)<sub>3</sub> (c) for Nd; Sm<sup>3+</sup> and Sm(OH)<sub>3</sub> (c) for Sm; Eu<sup>3+</sup>, Eu(OH)<sub>3</sub> (c) and Eu<sup>2+</sup> for Eu; Gd<sup>3+</sup> and Gd(OH)<sub>3</sub> (c) for Gd; PbO<sub>2</sub> (c), Pb(OH)<sub>2</sub> (c) and Pb (c) for Pb. The Eh and pH of the *in vitro* reactions were mapped (shown in the circle with the pink to red). The compositions of *in vitro* synthesized crystalline NMs using the cell extract at pH 7.5 according to the XRD results [β-Co(OH)<sub>2</sub>, β-Ni(OH)<sub>2</sub>, ZnO, β-Cd(OH)<sub>2</sub>, Cd(OH)<sub>2</sub>, La(OH)<sub>3</sub>, CeO<sub>2</sub>, Pr(OH)<sub>3</sub>, Nd(OH)<sub>3</sub>, Sm(OH)<sub>3</sub>, Eu(OH)<sub>3</sub>, Gd(OH)<sub>3</sub> and Pb<sub>3</sub>(NO)<sub>3</sub>(OH)<sub>5</sub> and Pb<sub>2</sub>(NO)<sub>3</sub>(OH)<sub>3</sub>], which are indicated by the yellow box.



**Fig. S23.** Pourbaix diagram analyses and measurements of the Eh and pH in the *in vivo* conditions of amorphous NMs or no NM formation at pH 7.5. (*A*) Formation of amorphous NMs at pH 7.5. The species taken into account include:  $SeO_4^{2^-}$ ,  $HSeO_3^{2^-}$ ,  $SeO_3^{2^-}$ , Se(c) and  $HSe^-$  for Se;  $Y^{3+}$  and  $Y(OH)_3(c)$  for Y;  $ZrO_2(c)$  for Zr;  $H_2TeO_4$ ,  $HTeO_4^-$ ,  $TeO(OH)_3^-$ ,  $TeO_2(OH)_2^{2^-}$ , Te (c) and  $Te_2^{2^-}$  for Te. (*B*) No NM formation pH at 7.5. The species taken into account include:  $B(OH)_3$  and  $H_2BO_3$  for B;  $Al(OH)_3$  (cr) for Al;  $VO_2(OH)_2^-$ ,  $VO_3OH^{2^-}$ ,  $V_6O_{13}(c)$ ,  $V_2O_4(c)$ ,  $V_3O_5(c)$ ,  $V_2O_3(c)$  and  $VOH^+$  for V;  $HCrO_4^-$ ,  $CrO_2(c)$ ,  $CrO_4^{2^-}$ ,  $Cr_2O_3(cr)$  and  $Cr^{2+}$  for Cr;  $MoO_4^{2^-}$ ,  $MoO_2(cr)$  and Mo (c) for Mo;  $Ba^{2+}$  and  $BaCO_3(c)$  for Ba. The Eh and pH of the *in vitro* reactions were mapped (shown in the circle with the pink to red).



**Fig. S24.** TEM images and EDS graphs of the Ni, Zn, Nd and Sm NMs synthesized *in vitro* at initial pH 7.5 for 30 min. (*A*) TEM images of the NMs synthesized *in vitro* with their corresponding elements labeled in orange circle. (*B*) The EDS graphs of the NMs synthesized *in vitro*. The NMs synthesized *in vitro* with their corresponding elements labeled in orange synthesized *in vitro* with their corresponding elements labeled *in vitro* with their corresponding elements.



**Fig. S25.** TEM images and EDS graphs of the 12 crystalline NMs synthesized *in vitro* at the pH 7.5. (*A*) TEM images of the crystalline NMs synthesized *in vitro* with their corresponding elements labeled in orange circle. Scale bar, 20 nm. (*B*) The EDS graphs of the crystalline NMs synthesized *in vitro*. The amorphous NMs synthesized *in vitro* with their corresponding elements labeled in orange square.



**Fig. S26.** HR-TEM images and the XRD patterns of 12 crystalline NMs synthesized *in vitro* at pH 7.5. (*A*) HR-TEM images of the crystalline NMs synthesized *in vitro* at pH 7.5 with their corresponding elements labeled in orange circle. The interplanar distance of the crystalline NM lattice and the Miller indices of crystallographic planes within parentheses are shown together. (*B*) The crystalline composition confirmation of the biosynthesized crystalline NMs were analyzed using XRD. The orange numbers in parentheses represent the JCPDS cards. The black numbers in parentheses represent Miller indices corresponding to each scattering peak.


**Fig. S27.** Reconstructed crystal structures of 13 biosynthesized crystalline NMs at initial pH 7.5. Illustrations of crystalline structures of the biosynthesized crystalline NMs at initial pH 7.5 based on the XRD data. Colored spheres and sticks represent individual atoms and chemical bonds between the atoms, respectively.



**Fig. S28.** TEM images and EDS graphs of the amorphous NMs synthesized *in vitro* at pH 7.5. (*A*) TEM images of the amorphous NMs synthesized *in vitro* with their corresponding elements labeled in gray circle. Scale bar, 20 nm. (*B*) The EDS graphs of the amorphous NMs synthesized *in vitro*. The amorphous NMs synthesized *in vitro* with their corresponding elements labeled in gray square.



**Fig. S29.** Pourbaix diagram analyses and measurements of Eh and pH during *in vivo* and *in vitro* biosynthesis of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM. (*A*) *In vivo* synthesis of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM. The Eh and pH of the *in vivo* reactions were mapped (shown in the circle with the pink to red). (*B*) *In vitro* synthesis of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM. The species taken into account include: Fe<sub>2</sub>O<sub>3</sub> (c), Fe<sub>3</sub>O<sub>4</sub> (c), Fe<sup>2+</sup>, Fe(OH)<sub>2</sub> (c) and Fe (c) for Fe. The Eh and pH of the *in vitro* reactions were mapped (shown in the circle with the pink to red). The compositions of biosynthesized NM according to the XRD result ( $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>) is indicated by the yellow box.



**Fig. S30.** TEM images and EDS analysis of α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH and α-FeOOH NMs synthesized *in vivo* and *in vitro*. TEM image (*A*; shown with 20 nm scale bar) and EDS graph (*B*; with their corresponding element labeled in brown square) of α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH and α-FeOOH NMs synthesized *in vivo*. TEM image (*C*; shown with 20 nm scale bar) and EDS graph (*D*; with their corresponding element labeled in light green square) of α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH and α-FeOOH NMs synthesized *in vivo*. TEM image (*C*; shown with 20 nm scale bar) and EDS graph (*D*; with their corresponding element labeled in light green square) of α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH and α-FeOOH NMs synthesized *in vitro*. TEM image (*E*; shown with 20 nm scale bar) and EDS graph (*F*; with their corresponding element after calcination labeled in brown square) of α-Fe<sub>2</sub>O<sub>3</sub> NM synthesized *in vivo* after calcination. TEM image (*G*; shown with 20 nm scale bar) and EDS graph (*H*; with their corresponding element after calcination labeled in brown square) of α-Fe<sub>2</sub>O<sub>3</sub> NM synthesized *in vivo* after calcination. TEM image (*G*; shown with 20 nm scale bar) and EDS graph (*H*; with their corresponding element after calcination labeled in light green square) of α-Fe<sub>2</sub>O<sub>3</sub> NM synthesized *in vivo* after calcination.



**Fig. S31.** HR-TEM images, XRD patterns and reconstructed crystal structures of the biosynthesized Fe NM. (*A*) HR-TEM image of α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH and α-FeOOH NMs synthesized *in vivo* by 6-hour reaction with the corresponding element labeled in brown circle. (*B*) XRD analysis of the α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH and α-FeOOH NMs synthesized *in vivo*. The brown numbers in parentheses represent the JCPDS cards. (*C*) HR-TEM image of the α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH and α-FeOOH NMs synthesized *in vivo*.

corresponding element labeled light green circle. (D) XRD analysis of the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> and  $\beta$ -FeOOH and  $\alpha$ -FeOOH NMs synthesized *in vitro*. The light green numbers in parentheses represent the JCPDS cards. (E) Illustrations of crystalline structures of the biosynthesized α-Fe<sub>2</sub>O<sub>3</sub> NM from XRD analysis data. (F) Illustrations of crystalline structures of the biosynthesized β-FeOOH NM from XRD analysis data. (G) HR-TEM image of in vivo synthesized  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM after calcination at 800°C. (H) XRD analysis of the *in vivo* synthesized  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM after calcination at 800°C. The brown numbers in parentheses represent the JCPDS card. (I) HR-TEM image of in vitro synthesized α-Fe<sub>2</sub>O<sub>3</sub> NM after calcination at 800°C. (J) XRD analysis of the *in vitro* synthesized α-Fe<sub>2</sub>O<sub>3</sub> NM after calcination. The light green numbers in parentheses represent the JCPDS card. (K) Illustrations of the crystalline structures of the biosynthesized crystalline  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM after calcination at 800°C from XRD analysis data. In the HR-TEM images, the interplanar distance of the lattice (indicated in yellow in A, C, G and I) and the Miller indices (shown in numbers in parentheses in B, D, H and J) of crystallographic planes within parentheses are shown together. In E, F and K, colored spheres and sticks represent individual atoms and chemical bonds between the atoms, respectively.



**Fig. S32.** Pourbaix diagrams for bi-elemental combinations resulting in multi-element crystalline NMs. Pourbaix diagrams of bi-elemental combinations predicted to form multi-element crystalline NMs. Those bi-elemental combinations tested for NM biosynthesis in this study are indicated in light dark blue square. Those bi-elemental combinations not tested are shown in gray square, while those NMs previously biosynthesized (8) are shown in light green square. The compositions of the multi-element crystalline NMs *in vivo* and *in vitro* according to the XRD results (Ag<sub>2</sub>S, CoFe<sub>2</sub>O<sub>4</sub>, NiFe<sub>2</sub>O<sub>4</sub>, ZnFe<sub>2</sub>O<sub>4</sub>, PbMoO<sub>4</sub>, Ag<sub>2</sub>WO<sub>4</sub> and PbWO<sub>4</sub>), which are indicated by the yellow box. (Fig. S32. continued)





**Fig. S33.** Pourbaix diagrams of those element combinations that could not be predicted to form bi-elements. These element combinations for the corresponding bi-elements shown in blue square did not exist in the database, and thus could not be predicted by Pourbaix diagram analyses. (Fig. S33. continued)





**Fig. S34.** Pourbaix diagrams of bi-elemental combinations for the multi-element crystalline NMs. (*A*) Pourbaix diagrams of bi-elemental combinations shown in red square for multi-element crystalline NMs synthesized *in vivo*. (*B*) Pourbaix diagrams of two separate single-element NMs synthesized *in vivo*. (*C*) Pourbaix diagrams of single-element NMs synthesized *in vivo*. The corresponding elements are shown in blue square (*B*, *C*). (Fig. S34. continued)



(Fig. S34. continued)





**Fig. S35.** TEM images and EDS graphs of the multi-element crystalline NMs synthesized *in vivo*. (*A*) TEM images of the multi-element crystalline NMs synthesized *in vivo* with their corresponding elements labeled in dark blue circle. Scale bar, 20 nm. (*B*) The EDS graphs of the multi-element crystalline NMs synthesized *in vivo*. The crystalline NMs synthesized *in vivo* with their corresponding elements labeled in dark blue circle. Scale bar, 20 nm. (*B*) The EDS graphs of the multi-element crystalline NMs synthesized *in vivo*. The crystalline NMs synthesized *in vivo* with their corresponding elements labeled in dark blue square.



**Fig. S36.** XRD patterns of multi-element crystalline NMs biosynthesized *in vivo* and *in vitro*. (*A*) The crystalline compositions of multi-element NMs synthesized *in vivo* at pH 6.5. The dark blue numbers in parentheses represent the JCPDS cards. Because cell debris existed for NMs synthesized *in vivo*, background scattering from the some organic

components was also recorded in the XRD patterns. (*B*) The crystalline compositions of multi-element NMs synthesized *in vivo* at pH 6.5. These NMs were predicted not producible from Pourbaix diagram analysis, but could be biosynthesized. The dark blue numbers in parentheses represent the JCPDS cards. (*C*) The crystalline compositions of multi-element NMs synthesized *in vitro* at pH 6.5. The light blue numbers in parentheses represent the JCPDS cards. (*D*) The crystalline compositions of Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM synthesized *in vivo* at pH 7.5. The dark blue numbers in parentheses represent the JCPDS cards. (*E*) The crystalline compositions of Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM synthesized *in vivo* at pH 7.5. The dark blue numbers in parentheses represent the JCPDS cards. (*E*) The crystalline compositions of Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM synthesized *in vitro* at pH 7.5. The black numbers in parentheses represent Miller indices corresponding to each scattering peak.



**Fig. S37.** TEM images of various single-element NMs synthesized *in vivo*. (*A*) TEM images of crystalline Ag NM synthesized *in vivo*. (*B*) TEM images of single amorphous NMs synthesized *in vivo*. (*C*) TEM images of two separate single-element NMs synthesized *in vivo*. (*D*) TEM images of single-element



**Fig. S38.** TEM images and EDS graphs of the multi-element crystalline NMs synthesized *in vivo*. These NMs were biosynthesized *in vivo* even though they were predicted not producible from Pourbaix diagram analysis. (A) TEM images of the multi-element crystalline NMs synthesized *in vivo* with their corresponding elements labeled in dark blue circle. (*B*) The EDS graphs of the multi-element crystalline NMs synthesized *in vivo* with their corresponding elements labeled *in vivo*. The crystalline NMs synthesized *in vivo* with their corresponding elements labeled *in vivo*. The synthesized *in vivo* with their corresponding elements labeled *in dark* blue square.



**Fig. S39.** TEM images and EDS graphs of multi-element crystalline NMs synthesized *in vitro*. (*A*) TEM images of multi-element crystalline NMs synthesized *in vitro* with their corresponding elements labeled in light blue circle. Scale bar, 20 nm. (*B*) The EDS graphs of multi-element crystalline NM synthesized *in vitro*. The crystalline NMs synthesized *in vitro* with their corresponding elements labeled in light blue square.



**Fig. S40.** TEM images of various single-element NMs synthesized *in vitro*. (*A*) TEM images of crystalline Ag NM synthesized *in vitro*. (*B*) TEM images of single amorphous NMs synthesized *in vitro*. (*C*) TEM images of two separate single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*.



**Fig. S41.** TEM images of various single-element NMs synthesized *in vivo* at pH 7.5. (*A*) TEM images of crystalline Ag NM synthesized *in vivo*. (*B*) TEM images of two separate single-element NMs synthesized *in vivo*. (*C*) TEM images of single-element NMs synthesized *in vivo*. Amorphous NMs are indicated with (a) next to the element name.



**Fig. S42.** TEM images of various single-element NMs synthesized *in vitro* at pH 7.5. (*A*) TEM images of single crystalline NMs synthesized *in vitro*. (*B*) TEM images of single amorphous Se NM synthesized *in vitro*. (*C*) TEM images of two separate single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*. (*D*) TEM images of single-element NMs synthesized *in vitro*.



**Fig. S43.** TEM images, EDS graphs and XRD patterns of V/Ag and Cr/Zn bi-elemental combinations resulting in formation of crystalline NMs *in vivo* and *in vitro* at pH 7.5. For each TEM result with indicated bi-elements, the left is the TEM image and the right is the HR-TEM image. TEM images (*A*) and EDS graph (*B*; with their corresponding element labeled in red square) of Na<sub>4</sub>VO<sub>2</sub>, Na<sub>3</sub>AgO<sub>2</sub> and Ag<sub>2</sub>O NMs synthesized *in vivo*. TEM images (*C*) EDS graph (*D*; with their corresponding element labeled in orange square) and XRD patterns (*E*) of Na<sub>4</sub>VO<sub>2</sub>, Na<sub>3</sub>AgO<sub>2</sub> and Ag<sub>2</sub>O NMs synthesized *in vitro*. The orange numbers in parentheses represent the JCPDS cards. The black numbers in parentheses represent Miller indices corresponding to each scattering peak. TEM images (*F*) and EDS graph (*G*; with their corresponding element labeled in red square) of Na<sub>4</sub>CrO<sub>4</sub> and Zn(OH)<sub>2</sub> NMs synthesized *in vivo*. TEM images (*H*) EDS graph (*I*; with their corresponding element labeled in orange square) and XRD patterns (*J*) of Na<sub>4</sub>CrO<sub>4</sub> and Zn(OH)<sub>2</sub> NMs synthesized *in vivo*.



**Fig. S44.** Pourbaix diagrams for V/Pb, and TEM images and EDS graphs of Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM synthesized *in vivo* and *in vitro* at pH 7.5. (*A*) Pourbaix diagrams for V/Pb during biosynthesis crystalline NMs. The predicted compositions of multi-element NMs taken into account include Pb<sub>3</sub>(VO<sub>4</sub>)<sub>2</sub> and Pb<sub>2</sub>V<sub>2</sub>O<sub>7</sub>. (*B*) TEM image of Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM synthesized *in vivo* with the corresponding elements labeled in red circle. (*C*) The EDS graph of Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM synthesized *in vivo* with the corresponding elements labeled in red square. (*D*) TEM image of Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM synthesized *in vivo* with the corresponding elements labeled in synthesized *in vitro* with the corresponding elements labeled in vitro with the corresponding elements labeled *in vitro* with the corresponding elements labeled *in orange* square.



**Fig. S45.** Reconstructed crystal structures of biosynthesized multi-element crystalline NMs. Illustrations of crystalline structures of the biosynthesized multi-element NMs based on the XRD data. Colored spheres and sticks represent individual atoms and chemical bonds between the atoms, respectively.



**Fig. S46.** HR-TEM images of crystalline Ag<sub>2</sub>TeO<sub>3</sub> NM synthesized *in vivo* and *in vitro*, and the correlation graphs between particle size and precursor concentration. HR-TEM images of crystalline Ag<sub>2</sub>TeO<sub>3</sub> NM synthesized *in vivo* using AgNO<sub>3</sub> and Na<sub>2</sub>TeO<sub>3</sub> at precursor concentrations of 0.25 and 1 mM each for both elements (*A*, *B*). HR-TEM image obtained with 0.5 mM precursors is shown in Fig. 3*A*. HR-TEM images of the crystalline Ag<sub>2</sub>TeO<sub>3</sub> NM synthesized *in vitro* using AgNO<sub>3</sub> and Na<sub>2</sub>TeO<sub>3</sub> at precursor concentrations of 0.5 mM precursors is shown in Fig. 3*A*. HR-TEM image obtained with 1 mM precursors is shown in Fig. 3B. The correlation graphs of the crystalline Ag<sub>2</sub>TeO<sub>3</sub> NMs synthesized *in vivo* at varying precursor concentrations. (*E*) The sizes of crystalline Ag<sub>2</sub>TeO<sub>3</sub> NMs synthesized *in vivo* with corresponding precursors of 0.25 mM each (13.69  $\pm$  0.87 nm), 0.5 mM each (18.90  $\pm$  1.02 nm) and 1 mM each (21.06  $\pm$  1.48 nm), respectively. (*F*) The correlation graphs of the crystalline Ag<sub>2</sub>TeO<sub>3</sub> NMs synthesized *in vitro* with corresponding precursors of 0.25 mM each (13.69  $\pm$  0.87 nm), 0.5 mM each (18.90  $\pm$  1.02 nm) and 1 mM each (21.06  $\pm$  1.48 nm), respectively. (*F*) The correlation graphs of the crystalline Ag<sub>2</sub>TeO<sub>3</sub> NMs synthesized *in vitro* at varying precursor concentrations. The sizes of crystalline Ag<sub>2</sub>TeO<sub>3</sub> NMs synthesized *in vitro* with corresponding precursors of 0.5 mM each (21.21  $\pm$  0.81 nm) and 2 mM each (34.96  $\pm$  1.38 nm), respectively.



**Fig. S47.** XRD patterns of the crystalline  $Ag_2TeO_3$  NM synthesized *in vivo* and *in vitro* at varying precursors concentrations. The black numbers in parentheses represent Miller indices corresponding to each scattering peak and matched with JCPDS database (JCPDS card. No. 46-0036). XRD patterns of crystalline  $Ag_2TeO_3$  NM synthesized *in vivo* with the corresponding precursors (0.25 mM each and 1 mM each, respectively) labeled in dark blue line. XRD patterns of crystalline  $Ag_2TeO_3$  NM synthesized *in vitro* with corresponding precursors (0.25 mM each and 1 mM each, respectively) labeled in light blue line.

In vivo synthesis



**Fig. S48.** Summary diagram showing the results of *in vivo* and *in vitro* biosynthesis of various single and multi-element NMs. Single-element NMs synthesized *in vivo* and *in vitro* are shown at the top (in red box) and on the left (in blue box), respectively. Bi-element NMs synthesized *in vivo* and *in vitro* are shown in upper triangle region (with red border) and lower triangle region (with blue border), respectively. NS, No NM synthesis; Amorphous NMs are indicated with (a) next to the element names. A larger image of this figure can be found in Dataset S2.



**Fig. S49.** FTIR graphs of the crystalline NMs synthesized *in vivo*. Numbers in graphs show wavenumbers (cm<sup>-1</sup>) that correspond to surface functional groups attached to crystalline NMs synthesized *in vivo*.



**Fig. S50.** FTIR graphs of the amorphous NMs synthesized *in vivo*. Numbers in graphs show wavenumbers (cm<sup>-1</sup>) that correspond to surface functional groups attached to amorphous NMs synthesized *in vivo*.



**Fig. S51.** FTIR graphs of the multi-element crystalline NMs synthesized *in vivo*. Numbers in graphs show wavenumbers (cm<sup>-1</sup>) that correspond to surface functional groups attached to crystalline multi-element NMs synthesized *in vivo*.



**Fig. S52.** FTIR graphs of the crystalline and amorphous NMs synthesized *in vitro*. (*A*) FTIR graphs of the crystalline NMs synthesized *in vitro*. Numbers in graphs show wavenumbers (cm<sup>-1</sup>) that correspond to surface functional groups attached to crystalline NMs synthesized *in vitro*. (*B*) FTIR graphs of the amorphous NMs synthesized *in vitro*. Numbers in graphs show wavenumbers (cm<sup>-1</sup>) that correspond to surface functional groups attached to amorphous NMs synthesized *in vitro*.



**Fig. S53.** FTIR graphs of the multi-element crystalline NMs synthesized *in vitro*. Numbers in graphs show wavenumbers (cm<sup>-1</sup>) that correspond to surface functional groups attached to crystalline multi-element NMs synthesized *in vitro*.



**Fig. S54.** Magnetic field-dependent magnetization curves (M-H curves) of magnetic NMs synthesized *in vivo* and *in vitro*. M-H curves indicate different magnetic properties of magnetic NMs synthesized *in vivo* and *in vitro*. Magentic moments of crystalline Mn<sub>3</sub>O<sub>4</sub> (*A*), Fe<sub>3</sub>O<sub>4</sub> (*B*), CoFe<sub>2</sub>O<sub>4</sub> (*C*), NiFe<sub>2</sub>O<sub>4</sub> (*D*), ZnMn<sub>2</sub>O<sub>4</sub> (*E*) and ZnFe<sub>2</sub>O<sub>4</sub> (*F*) NMs were measured under magnetic field strength of -50,000 to 50,000 Oe at 300 K. Each saturation magnetization (*Ms*) value of the biosynthesized magnetic NM is shown in the graphs.

## **SI Appendix Tables**

Strains or	Relevant characteristics	Reference
plasmids	Abbreviations: Km <sup>r</sup> , kanamycin resistance.	or source
Strains		
Escherichia coli	<i>E. coli</i> K-12 F <sup>-</sup> $\varphi$ 80 <i>lacZ</i> $\Delta$ <i>M15</i> $\Delta$ ( <i>lacZYA-argF</i> )	Invitrogen
DH5a	U169 recA1 endA1 hsdR17( $r_k m_k^+$ ) phoA supE44 thi- lowrA96 relA1 $\lambda^-$	
DH5a-nTac15K	$DH5\alpha$ containing nTac15K Km <sup>r</sup>	This study
Dilou prueron	brisa containing practors, ixin	This study
YJ-MT-PCS	DH5a containing pYJ-MT-PCS, Km <sup>r</sup>	This study
Plasmids		
pTac15K	pACYC177 derivative, p15A origin, tac promoter,	(9)
	rrnBT1T2 terminator, Km <sup>r</sup> (4.0 kb)	
pYJ-MT	p15A origin, tac promoter, rrnB T1 T2 terminator,	This study
	pTac15K derivative, Km <sup>1</sup> , MT gene from	
-VI DCC	<i>Pseudomonas putida</i> metallothionein (4.2 kb)	
prj-PCS	pT3A origin, <i>lac</i> promoter, rmB 11 12 terminator, pT3c15K derivative Km <sup>r</sup> PCS gene from	This study
	Arabidonsis thaliana ecotype Columbia (leaves)	
	(5.4 kb)	
pYJ-MT-PCS	p15A origin, double <i>tac</i> promoter, rrnB T1 T2	This study
1	terminator, pTac15K derivative, Km <sup>r</sup> , co-expression	5
	of MT and PCS gene (5.7 kb)	

## Table S1. Bacterial strains and plasmids used in this study

\* Abbreviations: Km, Kanamycin; r, resistance.

$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	In vivo synthesized NMs at initial pH 6.5			In vitro synthesized NMs at initial pH 6.5					
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Element	Chemical species	Size (nm)	Shape	Element	Chemical species	Size (nm)	Shape	
Fe         Fe/O         4.46 ± 0.27         Spherical         Fe         Fe/O         6.32 ± 1.47         Spherical           Se         Amorphous         105.25 ±         Spherical         Se         Amorphous         9.0 ± 0.25         Spherical           Y         Amorphous         4.10 ± 0.22         Spherical         Se         Amorphous         9.3.6 ± 1.20.5         Spherical           Za         Amorphous         4.10 ± 0.22         Spherical         Ag         Ag         1.137 ± 2.45         Spherical           Amorphous         1.43 ± 0.23         Spherical         Ag         Sn         SnO         7.7 ± 1.41         Spherical           Te         Te         Te         Te         Sn         SnO         5.17 ± 0.33         Spherical           Ca         Amorphous         1.43 ± 1.14         Spherical         Au         Au         Au         1.82 ± 8.32         Spherical           Nd         Amorphous         1.43 ± 2.20.33         Spherical         Spherical         Spherical         Au         Au         Au         Au         Au         Au         Su ± 2.44         Au         Au         Au         Au         Au         Su ± 2.44         Su ± 4.24         Spherical <td< td=""><td>Mn</td><td>Mn<sub>3</sub>O<sub>4</sub></td><td><math>3.93 \pm 1.29</math></td><td>Spherical</td><td>Mn</td><td>Mn<sub>3</sub>O<sub>4</sub></td><td><math>20.84 \pm 1.39</math></td><td>Square</td></td<>	Mn	Mn <sub>3</sub> O <sub>4</sub>	$3.93 \pm 1.29$	Spherical	Mn	Mn <sub>3</sub> O <sub>4</sub>	$20.84 \pm 1.39$	Square	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Fe	Fe <sub>3</sub> O <sub>4</sub>	$4.46 \pm 0.27$	Spherical	Fe	Fe <sub>3</sub> O <sub>4</sub>	$6.32 \pm 1.47$	Spherical	
SeeAmorphous $105.25 \pm5pherical16.05See Amorphous93.56 \pm 1.205SelphericalApplicalAA$	Cu	Cu <sub>2</sub> O	$17.69 \pm 3.23$	Spherical	Cu	Cu <sub>2</sub> O	$4.90 \pm 0.25$	Spherical	
016.0016.00OpticitalZr.Amorphous3.48ApplericalZr.Amorphous21.78 + 2.34ApplericalAg.Ag.1.87 + 2.36SphericalAnMolO:20.58 + 1.70RodSphericalInInO(1)31.69 + 2.35SquareAg.Ag.Ag.NoSphericalTeAmorphous21.58 + 2.16SphericalInSifty5.17 + 0.33SphericalTeAmorphous21.58 + 2.16SphericalLaAmorphous16.18 + 1.95SphericalAuAuAu18.24 + 2.34SphericalPrAmorphous16.18 + 1.95SphericalAuAuAu4.94 + 0.24SphericalRdAmorphous17.27 + 1.45SphericalCo $\beta - Ca(OH)$ $4.94 + 0.24$ SphericalRdAmorphous15.27 + 1.35SphericalSeAmorphous3.63 + 0.43SphericalAuAu20.06 + 2.27SphericalZrAmorphous3.63 + 0.43SphericalAuAu20.06 + 2.27SphericalZrAmorphous3.63 + 0.43SphericalAuAu20.06 + 2.27SphericalZrAmorphous3.74 + 0.23SphericalAuAu20.06 + 2.27SphericalZrAmorphous3.76 + 0.41ApplericalAuAu20.06 + 2.27SphericalZrAmorphous3.76 + 0.41ApplericalAuAu20.01 + 3.14Applerical </td <td>Se</td> <td>Amorphous</td> <td><math>105.25 \pm</math></td> <td>Spherical</td> <td>Se</td> <td>Amorphous</td> <td><math>93.36 \pm 12.05</math></td> <td>Spherical</td>	Se	Amorphous	$105.25 \pm$	Spherical	Se	Amorphous	$93.36 \pm 12.05$	Spherical	
YAmorphous4.10 ± 0.22SphericalZrAmorphous2.178 ± 2.44AgeMoMo(Y)20.58 ± 1.70RodAgAgAgShericalAgAgShericalShericalInIn(OII)3.82 ± 0.33SphericalAuAmorphous1.53 ± 1.14SphericalCeAmorphous1.43 ± 1.14SphericalCeAmorphous1.18 ± 1.95SphericalNdAmorphous1.18 ± 1.95SphericalNdAmorphous1.18 ± 1.95SphericalNdAmorphous1.18 ± 1.95SphericalRuAmorphous1.15 ± 1.20SphericalRuAuAuAuSphericalAuAuAuAuSphericalAuAuAuAuSphericalAuAuAuSphericalYAmorphous1.53 ± 1.20SphericalCo $\beta - C40(DH)$ Sc 2.1 ± 2.67PbAmorphous5.25 ± 0.28SphericalYAmorphousCo $\beta - C40(DH)$ Sc 1.1 ± 2.74RodIaIaLenemetChemical speciesSize(mi)ShapeCo $\beta - C40(DH)$ Sc 1.1 ± 7.35SeAmorphous2.3 ± 3.45SphericalSphericalRodGdGd (Gd(H)Size(m)ShapeCo $-Ca(O(H))$ Sc 1.2 ± 1.2 5.75SmSm(O(H)1.1 ± 1.71<	50	Amorphous	16.05	Spherical	Zr	Amorphous	$34.89 \pm 3.38$	Atypical	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Y	Amorphous	$4.10 \pm 0.22$	Spherical	Ag	Âg	$11.87 \pm 2.46$	Spherical	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Zr	Amorphous	$21.78 \pm 2.34$	Atypical	In	In(OH)3	$31.69 \pm 2.85$	Square	
Ag InAg In932 + 0.3 (11)Spherical Spherical 	Mo	MoO <sub>2</sub>	$20.58 \pm 1.70$	Rod	Sn	SnO <sub>2</sub>	$7.72 \pm 1.41$	Spherical	
In $(10(11))$ $3.82 \pm 0.33$ Spherical Pb Anorphous $4.24 \pm 3.24$ Spherical Pb Anorphous $4.24 \pm 3.24$ Spherical Pb Anorphous $4.24 \pm 3.24$ Spherical Pc Anorphous $4.24 \pm 3.24$ Spherical Pi Anorphous $4.24 \pm 3.24$ Spherical Nd Anorphous $12.15 \pm 1.55$ Spherical Spherical Sin Anorphous $11.56 \pm 1.75$ Spherical Ea Anorphous $11.56 \pm 1.75$ Spherical Nd Anorphous $15.35 \pm 1.55$ Spherical Nd Anorphous $4.74 \pm 3.24 + 2.04$ Rod, Plate Zn ZnO $6.62 \pm 0.43$ Spherical Nd Anorphous $15.35 \pm 1.55$ Spherical Nd Anorphous $4.74 \pm 3.26 + 3.24$ Nd Rod, Plate Zn ZnO $6.62 \pm 0.43$ Spherical Nd Anorphous $5.25 \pm 0.28$ Spherical Nd Anorphous $4.74 \pm 3.26 + 3.24$ Nd Spherical Nd Anorphous $5.25 \pm 0.28$ Spherical Nd Anorphous $3.71 \pm 2.74$ Nd Spherical Nd Anorphous $5.25 \pm 0.28$ Spherical Nd Anorphous $3.71 \pm 2.74$ Spherical Nd Anorphous $3.71 \pm 2.74$ Spherical Nd Anorphous $3.71 \pm 2.74$ Spherical Nd Nd Nd(OH), $5.71 \pm 2.67$ Rod, Plate Nd Nd Nd(OH), $5.71 \pm 2.67$ Rod, Plate Nd Nd Nd(OH), $5.73 \pm 3.24 + 3.24$ Rod, Price Co (Co(11)), CoO, $7.66 \pm 0.41$ Spherical La (La(0H)), $6.89 \pm 2.63$ Rod Se Anorphous $4.74 \pm 3.26 + 3.24$ Nd Nd Nd(OH), $1.774 \pm 3.26 + 3.24$ Rod, Price Co (Co(11)), CoO, $7.66 \pm 0.41$ Spherical La (La(0H)), $6.89 \pm 2.63$ Rod Sm Nd (Nd(OH)), $1.74 + 3.26 + 3.24$ Nd Nd (Nd(OH)), $1.794 \pm 3.26 + 3.24 + $	Ag	Ag	$9.86 \pm 1.96$	Spherical	Те	Amorphous	$21.58 \pm 2.16$	Spherical	
SnSn0; $5.17 + 0.43$ Spherical RodTeTeTe $5.49 \pm 9.16$ RodLaAmorphous14.31 \pm 1.14SphericalPAmorphous14.32 \pm 2.03SphericalPAmorphous14.32 \pm 2.03SphericalNiPAmorphous14.62 \pm 2.03SmSmorphous11.6 \pm 1.75GdAmorphous11.5 \pm 1.12GdAmorphous15.51 \pm 1.20AuAu20.06 \pm 2.27AuAu20.06 \pm 2.27SphericalSphericalPbAmorphous5.25 ± 0.28PbAmorphous5.25 ± 0.28PbAmorphous7.04 ± 0.39SphericalZrPhAmorphous7.04 ± 0.39SphericalZrAmorphous7.04 ± 0.39SphericalCcPoAmorphous7.04 ± 0.39SphericalZrAmorphous7.04 ± 0.39SphericalSphericalCoP-Ca(01h), Co,O1PoAmorphousCoP-Ca(01h), Co,O1SeAmorphousSeAmorphousSeAmorphousArAppicalCoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O1CoP-Ca(01h), Co,O	In	In(OH) <sub>3</sub>	$3.82 \pm 0.33$	Spherical	Au	Au	$18.24 \pm 3.24$	Spherical	
IcIc $33 \pm 1.14$ SphericalLaAmorphous16.18 ± 1.95SphericalCeAmorphous16.18 ± 1.95SphericalNdAmorphous12.7 ± 1.45SphericalSmAmorphous12.7 ± 1.45SphericalSmAmorphous15.5 ± 1.15SphericalSmAmorphous15.5 ± 1.25SphericalSphericalSeMuAmorphous15.5 ± 0.28PbAmorphous5.25 ± 0.28CoJCCOHI), CoO, 7.06 ± 0.41SphericalZrAmorphous15.15 ± 1.267RodPiCoJCCOHI), CoO, 7.06 ± 0.41SphericalSphericalNiJP-N(OH), 5.25 ± 0.25SphericalSphericalNiJP-N(OH), 5.25 ± 0.25SphericalSphericalNiJP-N(OH), 5.25 ± 0.25SphericalSphericalNiJP-N(OH), 5.42 ± 2.43SphericalSphericalCoJCCOHI), CoO, 4.09 ± 0.15SphericalSphericalCaAmorphousCoJCCOHI), CoO, 4.09 ± 0.15SphericalPiPrPr(OH), 5.24 ± 0.33RodGdGdGd(OH), 1.744 ± 1.05 <td>Sn</td> <td>SnO<sub>2</sub></td> <td><math>5.17 \pm 0.33</math></td> <td>Spherical</td> <td>Pb</td> <td>Amorphous</td> <td><math>4.94 \pm 0.24</math></td> <td>Spherical</td>	Sn	SnO <sub>2</sub>	$5.17 \pm 0.33$	Spherical	Pb	Amorphous	$4.94 \pm 0.24$	Spherical	
La Amorphous 14.81 = 1.45 Spherical In the probability of the probabi	le	le A sur sur la sur	$35.49 \pm 9.16$	Rod		In witho synthesized	NMs at initial pH 7.5		
CeAntorphous10 is 1.93SphericalNdAmorphous12.741.45SphericalNdAmorphous1.271.4.15SphericalSmAmorphous1.521.5SphericalEuAmorphous1.521.5SphericalGdAmorphous1.521.5SphericalAuAu20.062.27SphericalAuAu20.062.27SphericalPDAmorphous7.044.03SphericalPDAmorphous7.044.03SphericalPDAmorphous7.044.04SphericalPDAmorphous7.044.04SphericalCoP-C(OIII), CoO, 17.144.04NiP-C(OIII), CoO, 11.511.7SquareSeAmorphous3.43NiP-C(OIII), CoO, 11.511.7SquareSeAmorphous4.53RodSe2.50.23SeAmorphous2.951.8Anorphous2.951.122.07Rod1.512.07RodPiPiP(OII), 11.14CeCe(OII), CoO, 11.142.04CeCe(OII), CoO, 11.142.04SeAmorphous5.151.2ApricalAmorphous5.15SmSn(OII)1.12RodGdGd(OII)GdGd(OII)1.17Ru <td>La</td> <td>Amorphous</td> <td><math>14.31 \pm 1.14</math> 16.18 <math>\pm</math> 1.05</td> <td>Spherical</td> <td><b>F1</b> (</td> <td></td> <td></td> <td>01</td>	La	Amorphous	$14.31 \pm 1.14$ 16.18 $\pm$ 1.05	Spherical	<b>F1</b> (			01	
r Maraphous 1: 12: 52: 14: 35 Spherical Signerial Signerial Amorphous 1: 16: 17: 55 Spherical Signerial Signer Signerial Sign	Dr.	Amorphous	$10.18 \pm 1.93$ $14.82 \pm 2.03$	Spherical	Element	Chemical species	Size (nm)	Shape	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Nd	Amorphous	$14.02 \pm 2.05$ 12.75 + 1.45	Spherical	Co	β-Co(OH) <sub>2</sub>	$4.01 \pm 0.37$	Spherical	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Sm	Amorphous	$12.75 \pm 1.45$ 11.16 + 1.75	Spherical	N1	β-N1(OH) <sub>2</sub>	$28.28 \pm 4.49$	Rod, Plate	
LeaInterpretationSeeAmorphous $47/43 \pm 36.32$ ApprecialGdAmorphous $15.33 \pm 1.20$ SphericalYAmorphous $5.35 \pm 0.43$ SphericalAuAu $20.06 \pm 2.27$ SphericalZrAmorphous $6.65 \pm 0.41$ ApprecialHgAmorphous $7.04 \pm 0.39$ SphericalZrAmorphous $6.65 \pm 0.41$ ApprecialPbAmorphous $7.04 \pm 0.39$ SphericalCd $Gd$ $Gd(OH)_1$ $5.11 \pm 2.67$ Rod, PlateCo $\beta$ -N(iOH)_1 $0.51 \pm 1.2$ $2.67$ Rod, PlateEuLaLa(OH)_1 $80.58 \pm 2.63$ RodSeAmorphous $23.14$ AtypicalEuEu(OH)_1 $6.89 \pm 0.57$ RodGdGd(OH)_1 $10.18 \pm 1.71$ SquareSmSm(OH)_1 $17.83 \pm 4.33$ RodGdGd(OH)_1 $2.94 \pm 0.57$ RodRodPbNdNd(OH)_1 $80.58 \pm 2.63$ RodYAmorphous $2.55 \pm 0.23$ SphericalPbPbNd(OH)_1 $80.98 \pm 0.57$ RodGdGd(OH)_1 $5.47 \pm 0.37$ SphericalPbNd(OH)_1 $20.457 \pm 43.86$ PlatePrPr(OH)_1 $6.72 \pm 0.31$ SphericalPbNd(OH)_1 $10.38 \pm 2.04$ SphericalRodNdNd(OH)_1 $5.47 \pm 0.57$ RodFe $-6.7600$ $7.44 \pm 0.17$ SphericalRodGdGd(OH)_1 $5.47 \pm 0.57$ RodFe $-6.7600$ $7.44 \pm 0.17$	Fu	Amorphous	$15.77 \pm 1.75$	Spherical	Zn	ZnO	$6.62 \pm 0.35$	Spherical	
Gu Cu Hg Hg Amorphous2006 $\pm$ 2.27 Spherical Spherical Spherical SphericalY AmorphousAmorphous 6.5 $\pm$ 0.41Spherical Rod Cd Gd FC(O(H), 2.51,1 $\pm$ 7.40Rod Rod Rod Rod CoIm viro synthesized NNs at initial pH 7.5Im viro synthesized NNs at initial pH 7.5Im CoIm 	Gd	Amorphous	$15.27 \pm 1.33$ $15.53 \pm 1.20$	Spherical	Se	Amorphous	$447.43 \pm 36.32$	Atypical	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Δu	An	$10.05 \pm 1.20$ 20.06 ± 2.27	Spherical	Y	Amorphous	$5.36 \pm 0.43$	Spherical	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Hø	Amorphous	$5.25 \pm 0.28$	Spherical	Zr	Amorphous	$6.65 \pm 0.41$	Atypical	
Term <th cols<="" td=""><td>Ph</td><td>Amorphous</td><td><math>7.04 \pm 0.39</math></td><td>Spherical</td><td>Cd</td><td>β-Cd(OH)<sub>2</sub></td><td><math>54.71 \pm 7.40</math></td><td>Rod</td></th>	<td>Ph</td> <td>Amorphous</td> <td><math>7.04 \pm 0.39</math></td> <td>Spherical</td> <td>Cd</td> <td>β-Cd(OH)<sub>2</sub></td> <td><math>54.71 \pm 7.40</math></td> <td>Rod</td>	Ph	Amorphous	$7.04 \pm 0.39$	Spherical	Cd	β-Cd(OH) <sub>2</sub>	$54.71 \pm 7.40$	Rod
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		In vive synthesized N	Me at initial nH 7	5	Te	Amorphous	$30.71 \pm 2.74$	Spherical	
		In vivo synthesizeu N	vis at initial pri /		La	La(OH) <sub>3</sub>	$32.11 \pm 1.57$	Rod	
Co $\beta$ -Co(OH), Co(A) $A06 \pm 0.41$ Spherical       Pr       P(OH)) $59.25 \pm 5.30$ Rod         Zn       ZnO $10.51 \pm 1.71$ Square       Sm       Sm       Sm(OH)) $173.83 \pm 4.53$ Rod         Se       Amorphous $24.34$ Atypical       Sm       Sm (OH)) $173.83 \pm 4.53$ Rod         Y       Amorphous $2.96 \pm 0.12$ Atypical       Sm       Sm(OH)) $1.794.08 \pm 21.02$ Rod         Cd $\beta$ -Cd(OH), $5.02 \pm 0.15$ Spherical       Pb       Pb(NO)/(OH), $204.57 \pm 43.86$ Plate         Ce       Cc(OH), $6.02 \pm 0.93$ Rod       Rod       Ee $e-CO_1$ $Pb(NO)/(OH)$ $0.38 \pm 2.04$ Spherical         Pr       P(OH) $6.13 \pm 4.17$ Square       Spherical       In viro synthesized Fe NM for 6-hour incubation       Element       Chemical species       Size (nm)       Shape         Sm       Sm(OH) $15.47$ Rod       Rod       Fe $-FeOOH$ $0.38 \pm 2.04$ Spherical         Pb       Amorphous $56.15 \pm 6.17$ Rod       Rod       Rod       Fe $-FeOOH$ $7.69 \pm 0.51$	Element	Chemical species	Size(nm)	Shape	Ce	CeO <sub>2</sub>	$32.95 \pm 1.81$	Rod	
Nip-Ni(OH)22.3.11 $\pm 2.6$ Kod, PlateZnZnZnNdNd(OH)380.58 $\pm 2.63$ RodSeAmorphous187.51 $\pm$ AtypicalSmSm(OH)3173.83 $\pm 4.53$ RodYAmorphous2.96 $\pm 0.12$ AtypicalGdGd(OH)3 $(794.08 \pm 21.02)$ RodZrAmorphous2.96 $\pm 0.12$ AtypicalGdGd(OH)3 $(24.57 \pm 4.3.66)$ PlateCd $\beta$ -Cd(OH)2 $4.16 \pm 0.37$ SphericalPbPb <sub>2</sub> (NO) <sub>1</sub> (OH)2 $24.97 \pm 4.3.66$ PlateLaLa(DH)3 $28.99 \pm 1.87$ RodPbPb <sub>2</sub> (NO) <sub>1</sub> (OH)3 $204.57 \pm 4.3.66$ PlatePcCeCe(OH)3 $6.29 \pm 0.47$ SphericalPbPb <sub>2</sub> (NO) <sub>1</sub> (OH)3 $204.57 \pm 4.3.66$ PlatePrPt(DH)3 $67.22 \pm 0.93$ RodFe $\alpha$ -Feco,3, PFeOOH $10.38 \pm 2.04$ SphericalPdMo(OH)3 $11.77 \pm$ RodIn viro synthesized Fe NM for 6-hour cutivationIm viro synthesized multi-element crystalline NMs at initial PHElementChemical speciesSize (nm)ShapeSphericalPe $\alpha$ -FecO3, $\beta$ -FeOOH, $9.74 \pm 1.19$ SphericalPbAmorphous $56.1 \pm 6.17$ AtypicalAg, TPcCo $6.87 \pm 0.42$ SphericalPe $\alpha$ -FeOOH $9.74 \pm 1.21$ SphericalPe $\alpha$ -FeOOH $9.74 \pm 1.24$ SphericalPeCo $6.87 \pm 0.42$ SphericalAg, TAg, TAg	Co	β-Co(OH) <sub>2</sub> , Co <sub>3</sub> O <sub>4</sub>	$7.06 \pm 0.41$	Spherical	Pr	Pr(OH) <sub>3</sub>	$59.25 \pm 5.30$	Rod	
ZnZnO10.51 $\pm$ 1.71SquareSmSm(OH)3173.83 $\pm$ 4.53RodSeAmorphous4.55 $\pm$ 0.23SphericalEuEu(OH)36.89 $\pm$ 0.57RodZrAmorphous2.96 $\pm$ 0.12AtypicalGdGd(OH)31.794.08 $\pm$ 21.02RodCd $\beta$ -Cd(OH);4.16 $\pm$ 0.37SphericalPbPb <sub>2</sub> (NO);(OH)3204.57 $\pm$ 43.86PlateDataLaLa(OH)55.02 $\pm$ 0.15SphericalPbPb <sub>2</sub> (NO);(OH)3204.57 $\pm$ 43.86PlatePrPr(OH)56.02 $\pm$ 0.93RodPePbPb <sub>2</sub> (NO);(OH)32.04SphericalPrPr(OH)56.744 $\pm$ 5.19RodPePbPbNational speciesSize (nm)ShapeEuEu(OH)56.12 $\pm$ 0.77RodPePbPbNational speciesSize (nm)ShapeEuEu(OH)56.52 $\pm$ 0.93RodPaNatinital PH 6.5ElementCherical speciesSize (nm)ShapeFe $\alpha$ -FeOH36.51 $\pm$ 6.17AtypicalAtypicalAg, SAg, SAg, SAg, SSphericalPbAmorphous5.61 $\pm$ 6.17AtypicalAg, SAg, SAg, SAg, SSphericalRedChernical speciesSize (nm)ShapeShapeAg, SAg, S <td>N1</td> <td><math>\beta</math>-Ni(OH)<sub>2</sub></td> <td><math>25.11 \pm 2.67</math></td> <td>Rod, Plate</td> <td>Nd</td> <td>Nd(OH)<sub>3</sub></td> <td><math>80.58 \pm 2.63</math></td> <td>Rod</td>	N1	$\beta$ -Ni(OH) <sub>2</sub>	$25.11 \pm 2.67$	Rod, Plate	Nd	Nd(OH) <sub>3</sub>	$80.58 \pm 2.63$	Rod	
SeAmorphous $137.31 \pm \\ 24.34$ AtypicalEuEu (H)(h) $6.89 \pm 0.57$ RodYAmorphous $2.95 \pm 0.23$ SphericalGdGd(O(H) $1.794.08 \pm 21.02$ RodZrAmorphous $2.96 \pm 0.12$ AtypicalGdGd(O(H) $2.04.57 \pm 43.86$ PlateCd $\beta$ -Cd(O(H) $4.16 \pm 0.37$ SphericalPbPb <sub>2</sub> (NO)(O(H) $204.57 \pm 43.86$ PlateLaLa(OH) $2.8.99 \pm 1.87$ RodElementChemical speciesSize (nm)ShapePrPr (H) $6.1.9 \pm 4.17$ SquareFe $\alpha$ -FeOOH $0.38 \pm 2.04$ SphericalNdNd(OH) $61.19 \pm 4.17$ SquareFe $\alpha$ -FeOOH $0.38 \pm 2.04$ SphericalGdGd(O(H) $5.47 \pm 0.93$ RodFe $\alpha$ -FeOOH $0.38 \pm 2.04$ SphericalGdGd(O(H) $5.94 \pm 0.50$ RodFe, CoCoFeOOH $9.28 \pm 0.47$ SphericalFe $\alpha$ -FeOOH $5.51 - 17$ AtypicalFe, CoCoFeO4 $7.69 \pm 0.51$ SphericalFe $\alpha$ -FeOOH $9.74 \pm 1.19$ SphericalFe, ZaZnFeOO $7.45 \pm 1.07$ SphericalIn vivo synthesized Fe NM for 6-hour cultivationElementChemical speciesSize (nm)ShapeFe $\alpha$ -FeOOH $9.74 \pm 1.91$ SphericalAg, WAg, SAg, S<	Zn	ZnO	$10.51 \pm 1./1$	Square	Sm	Sm(OH) <sub>3</sub>	$173.83 \pm 4.53$	Rod	
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1Aniophous2.95 $\pm$ 0.12Atypical AtypicalPbPb(No)(OH)3, PbS(NO)(OH)3,204.57 $\pm$ 43.86PlateCd $\beta - Cq(OH)_1$ 4.16 $\pm$ 0.37Spherical SphericalIn viro synthesized Fe NM for 6-hour incubationLaLa(OH)_128.99 $\pm$ 1.87RodCcCe(OH)_1, CeO_24.09 $\pm$ 0.31Spherical SphericalPrPr(OH)_367.14 $\pm$ 5.19RodNdNd(OH)_361.19 $\pm$ 4.17SquareFuaEu(OH)_363.19 $\pm$ 4.03GdGd(OH)_35.49 $\pm$ 0.50RodGdGd(OH)_35.49 $\pm$ 0.50RodFeaFe(OD)7.46 $\pm$ 1.07BhAmorphous6.01 $\pm$ 0.58SphericalPhPhMoO_49.28 $\pm$ 0.47SphericalFe, CoCoFeO_47.69 $\pm$ 0.51PbAmorphous5.61.7AtypicalFeaFe/CO, BFe/CO7.69 $\pm$ 0.51SphericalFe/CO9.74 $\pm$ 1.19SphericalFeaFe/CO, B9.74 $\pm$ 1.19SphericalFeaFe/CO, B6.87 $\pm$ 0.42SphericalAg, TeAg: TeAg: TeAg: TeAg: TeAg, TeAg: SAggS13.47 $\pm$ 1.21SphericalFe, NiNiFe;Ou, BFe/OOH6.32 $\pm$ 0.54SphericalAg, TeAg: TeAg: TeAg: TeAg: TeAg, TeAg: TeAg: TeAg: TeAg: TeAg, TeAg: Te	v	Amorphous	24.54	Spharical	Gd	Gd(OH) <sub>3</sub>	$1,794.08 \pm 21.02$	Rod	
LaIntropartical p-Cd(OH)21.61 ± 0.37 spherical BaSpherical BaIntropartical prior Spherical PrPr(OH)2 Pr(OH)3Column Spherical Spherical PrIntropartical pr(OH)3Intropartical Spherical PrPrPr(OH)3 $6.24 \pm 5.19$ SmRod SmSm(OH)3 $11.17 \pm$ 1.547Rod PrSmSm(OH)3 $11.17 \pm$ 1.547Rod Spherical Gd $Gd(OH)3$ $5.47$ Spherical PbRod Amorphous $6.11 \pm 5.47$ Spherical PbEuEu(OH)3 $6.52 \pm 0.93$ Spherical PbRod Gd(OH)3 $5.47 \pm 0.30$ Spherical Pb $n vitro synthesized multi-element crystalline NMs at initial pHElementChemical speciesSize (nm)ShapeIn vitro synthesized Fe NM for 6-hour cultivationElementChemical speciesSize (nm)ShapeFe\alpha-FeOOH9.74 \pm 1.19SphericalPbShapeIn vitro synthesized multi-element crystalline NMs at initial pHre-6.5ElementChemical speciesSize (nm)ShapeSize (nm)ShapeMn, ZnPo, NiFe20a, β-FeOOH6.87 \pm 0.42SphericalFe, XnSphericalSphericalSquarefe, Ni NiFe20a, β-FeOOH6.87 \pm 0.42SphericalFe, XnSphericalSquarefe, XnPo BWOO49.91 \pm 0.46SphericalFe, XnSphericalSquarefe, ZnAg, SAg, SAg, SAg, SAg, SN AgesWo12.67 \pm 4.28SphericalAg, TeAg, TeAg, TeAg, TeAg, TeAg, TeA$		Amorphous	$4.33 \pm 0.23$ 2.96 ± 0.12	Atypical	Pb	$Pb_3(NO)_3(OH)_5$ ,	$204.57 \pm 43.86$	Plate	
In vitro synthesized Fe NM for 6-hour incubationIn vitro synthesized Fe NM for 6-hour cultivationIn vitro synthesized Fe NM for 6-hour cultivationIn vitro synthesized Fe NM for 6-hour cultivationIn vitro synthesized multi-element crystalline NMs at initial pHIn vitro synthesized multi-element crystalline NMs at initial pHCelementChemical speciesSize (nm)ShapeIn vivo synthesized multi-element crystalline NMs at initial pHCelementChemical speciesSize (nm)ShapeIn vivo synthesized multi-element crystalline NMs at initial pHCelementChemical speciesSize (nm)ShapeIn vivo synthesized multi-element crystalline NMs at initial pHCelementChemical speciesSize (nm)ShapeAg, SAg: SAg: SAg: S <th colspan<="" td=""><td>Cd</td><td>B-Cd(OH)</td><td><math>4.16 \pm 0.12</math></td><td>Spherical</td><td></td><td><math>Pb_2(NO)_3(OH)_3</math></td><td></td><td></td></th>	<td>Cd</td> <td>B-Cd(OH)</td> <td><math>4.16 \pm 0.12</math></td> <td>Spherical</td> <td></td> <td><math>Pb_2(NO)_3(OH)_3</math></td> <td></td> <td></td>	Cd	B-Cd(OH)	$4.16 \pm 0.12$	Spherical		$Pb_2(NO)_3(OH)_3$		
LaLaLa(OH); La(OH); CeC $28.99 \pm 1.87$ RodCeCe(OH); CeC $4.09 \pm 0.31$ PrSphericalFe $a-FeO3$ , $B-FeOH$ $10.38 \pm 2.04$ SphericalPrPr $11/17 \pm$ Rod $a-FeO3$ , $B-FeOH$ $10.38 \pm 2.04$ SphericalSmSm(OH); 	Ba	BaCO <sub>3</sub>	$5.02 \pm 0.15$	Spherical		In vitro synthesized Fe NM for 6-hour incubation			
CeCe (OH) <sub>3</sub> , $\hat{CeO}_2$ 4.09 ± 0.31Spherical RodPrPr(OH)_367.44 ± 5.19RodNdM(OH)_361.19 ± 4.17SquareSmSm(OH)_31.19 ± 4.17RodEuEu(OH)_36.52 ± 0.93RodGdGd(OH)_35.49 ± 0.50RodHgAmorphous56.15 ± 6.17AtypicalPbAmorphous56.15 ± 6.17AtypicalFe $\alpha$ -FeOOH9.74 ± 1.19SphericalFe $\alpha$ -FeOOH9.74 ± 1.19SphericalFe $\alpha$ -FeOOH9.74 ± 1.19SphericalFe $\alpha$ -FeOOH9.74 ± 1.19SphericalGeCoFeOH9.74 ± 1.19SphericalFe $\alpha$ -FeOOH9.74 ± 1.19SphericalAg. TeAg:TeAg:TeAg:TeAg:TeCoFeOH6.83 ± 0.45SphericalFe, ZnCoFeOH6.83 ± 0.45SphericalAg, TeAg:TeOCoFe:O46.83 ± 0.45SphericalFe, NiNiFe2O4,6.87 ± 0.42SphericalSphericalSphericalFe, ZnCoFe:O46.83 ± 0.45Ag:TeAg:TeO318.90 ± 1.02SphericalSphericalFe, ZnCoFe:O46.83 ± 0.45SplaricalSphericalFe, ZnCoFe:O46.83 ± 0.45SplaricalSphericalFe, ZnCoFe:O46.83 ± 0.45SplaricalSphericalFe, ZnCoFe:O46.83 ± 0.45Sp	La	La(OH)3	$28.99 \pm 1.87$	Rod	Element	Chemical species	Size (nm)	Shape	
Pr $Pr(OH)_3$ $67.44 \pm 5.19$ Rod $a \cdot FcOOH$ $10.38 \pm 2.04$ SphericalNdNd(OH)_3 $61.19 \pm 4.17$ SquareSmSm(OH)_3 $171.77 \pm$ RodEuEu(OH)_3 $6.52 \pm 0.93$ RodGdGd(OH)_3 $5.49 \pm 0.50$ RodHgAmorphous $6.01 \pm 0.58$ SphericalPbAmorphous $5.615 \pm 6.17$ AtypicalPbAmorphous $5.615 \pm 6.17$ AtypicalFe $a \cdot FeOOH$ $9.74 \pm 1.19$ SphericalFe $a \cdot FeOOH$ $9.74 \pm 1.19$ SphericalFe $a \cdot FeOOH$ $9.74 \pm 1.19$ SphericalAg. TeAg: TeAg: TeAg: TeAg: TeAg. NZn/MnO <sub>3</sub> $6.87 \pm 0.42$ SphericalFe, CoCoFe:OA $6.87 \pm 0.42$ SphericalFe, CoCoFe:OA $6.87 \pm 0.42$ SphericalFe, CoCoFe:OA $6.87 \pm 0.42$ SphericalFe, Ni <nife:oa, <math="">\beta -FeOOH<math>6.23 \pm 0.54</math>SphericalFe, CoCoFe:OA<math>6.87 \pm 0.42</math>SphericalFe, Ni<nife:oa, <math="">\beta -FeOOH<math>6.23 \pm 0.54</math>SphericalFe, Ni<nife:oa, <math="">\beta -FeOOH<math>6.23 \pm 0.54</math>SphericalFe, Ni<nife:oa, <math="">\beta -FeOOH<math>6.23 \pm 0.54</math>SphericalFe, ZoZn/MoO3<math>6.87 \pm 0.42</math>SphericalFe, Ni<nife:oa, <math="">\beta -FeOOH<math>6.23 \pm 0.54</math>SphericalFe, CoCoFe:OA<math>6.83 \pm 0.45</math>SphericalFe, ZiZn/Fe:OA<math>8.59 \pm 0.51</math>Spherical<!--</td--><td>Ce</td><td>Ce(OH)<sub>3</sub>, CeO<sub>2</sub></td><td><math>4.09 \pm 0.31</math></td><td>Spherical</td><td>Fe</td><td>α-Fe<sub>2</sub>O<sub>3</sub>, β-FeOOH,</td><td>10.29 + 2.04</td><td>Calcalized</td></nife:oa,></nife:oa,></nife:oa,></nife:oa,></nife:oa,>	Ce	Ce(OH) <sub>3</sub> , CeO <sub>2</sub>	$4.09 \pm 0.31$	Spherical	Fe	α-Fe <sub>2</sub> O <sub>3</sub> , β-FeOOH,	10.29 + 2.04	Calcalized	
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Gd $Gd(O(H)_3$ $5.49 \pm 0.50$ KodIn visco $10 \pm 0.51$ $10 \pm 0.51$ $5pherical$ HgAmorphous $6.01 \pm 0.58$ Spherical $Fe, Ni$ $NiFe_2O_4, \beta$ $FeOOH$ $7.45 \pm 1.07$ SphericalPbAmorphous $56.15 \pm 6.17$ Atypical $Fe, Ni$ $NiFe_2O_4, \beta$ $FeOOH$ $7.45 \pm 1.07$ SphericalElementChemical speciesSize (nm)Shape $Ag, S$ $Ag_2S$ $13.47 \pm 1.21$ SphericalFe $\alpha$ -FeOOH $9.74 \pm 1.19$ Spherical $Ag, S$ $Ag_2WO_4$ $7.56 \pm 1.02$ SphericalIn vivo synthesized multi-element crystalline NMs at initial pH $6.5$ $Ag, W$ $Ag_2WO_4$ $7.56 \pm 1.02$ SphericalElementChemical speciesSize (nm)Shape $W, Pb$ $PbWO_4$ $42.38 \pm 4.81$ SphericalFe, NiNiFe <sub>2</sub> O <sub>4</sub> , $\beta$ -FeOOH $6.87 \pm 0.42$ Spherical $Ag, W$ $Ag_2WO_4$ $7.56 \pm 1.02$ SphericalFe, NiNiFe <sub>2</sub> O <sub>4</sub> , $\beta$ -FeOOH $6.87 \pm 0.42$ Spherical $Ag, W$ $Ag_2TeO_5$ $8.45 \pm 0.61$ SphericalFe, NiNiFe <sub>2</sub> O <sub>4</sub> , $\beta$ -FeOOH $6.23 \pm 0.54$ Spherical $Square$ $Npherical$ $Ag, W$ $Ag_2TeO_3$ $8.90 \pm 1.02$ SphericalFe, NiNiFe <sub>2</sub> O <sub>4</sub> , $\beta$ -FeOOH $1.03 \pm 1.94$ Spherical $Square$ $NphericalSguareAg, SAg_2S10.93 \pm 1.94SphericalSphericalNphericalNphericalAg, WAg_2WO_412.67 \pm 4.28Spherical$	Eu	Eu(OH) <sub>3</sub>	$6.52 \pm 0.93$	Rod	Fe Co	CoFe <sub>2</sub> O <sub>4</sub>	$7.69 \pm 0.51$	Spherical	
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$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Fe	α-Fe <sub>2</sub> O <sub>3</sub> , β-FeOOH,	$9.74 \pm 1.10$	Spherical	Ag. Te	Ag <sub>2</sub> TeO <sub>3</sub>	$21.21 \pm 0.81$	Spherical	
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MO, PD         PDMOQ4 $9.91 \pm 0.46$ Spherical, Square           Ag, S         Ag <sub>2</sub> S $10.93 \pm 1.94$ Spherical           Ag, Te         Ag <sub>2</sub> TeO <sub>3</sub> $18.90 \pm 1.02$ Spherical           Ag, W         Ag <sub>2</sub> WO <sub>4</sub> $12.67 \pm 4.28$ Spherical           W, Pb         PbWO <sub>4</sub> $18.61 \pm 3.59$ Spherical           Hg, Te         Hg <sub>3</sub> TeO <sub>6</sub> $14.09 \pm 2.74$ Spherical           Hr vivo synthesized multi-element crystalline NM at initial pH         7.5           Elements         Chemical species         Size (nm)         Shape           V, Pb         Pb <sub>5</sub> (VO <sub>4</sub> ) <sub>3</sub> OH $9.60 \pm 1.46$ Spherical	Fe, Zn	ZnFe <sub>2</sub> O <sub>4</sub>	$8.45 \pm 0.61$	Spherical					
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V, Pb Pb <sub>5</sub> (VO <sub>4</sub> ) <sub>3</sub> OH $9.60 \pm 1.46$ Spherical	Elements	Chemical species	Size (nm)	Shape	•				
A second s	V. Ph	Pb5(VO4)3OH	$9.60 \pm 1.46$	Spherical					
	.,								

Table S2. Characteristics of NMs biosynthesized in vivo and in vitro
Primers	Sequences*(5'→3')		
P1	ATA <u>GAATTC</u> ATGAACGATCAACGCTGCGC		
P2	TAT <u>CTGCAG</u> TCAGGGCGAGATCGGATCACTC		
P3	ATA <u>GAATTC</u> ATGGCTATGGCGAGTTTATATCGG		
P4	TAT <u>GCATGC</u> TTAATAGGCAGGAGCAGCGAGATC		
P5	ATA <u>CTGCAG</u> TTGACAATTAATCATCGGCTCGTATA		
P6	P6 TAT <u>GCATGC</u> TTAATAGGCAGGAGCAGCGAGA		

Table S3. Oligo nucleotides used for PCR amplification in this study

\*Underlines indicate restriction enzyme sites.

No.	Elements	Elements category	Chemical types
1	Li	Alkali metal	CH <sub>3</sub> COOLi · 2H <sub>2</sub> O
2	В	Metalloid	H <sub>3</sub> BO <sub>3</sub>
3	Al	Post-transition metal	AlCl <sub>3</sub> ·6H <sub>2</sub> O
4	S	Nonmetal	$Na_2S$
5	V	Transition metal	NaVO <sub>3</sub>
6	Cr	Transition metal	CrCl <sub>3</sub> ·6H <sub>2</sub> O
7	Mn	Transition metal	MnCl <sub>2</sub> ·4H <sub>2</sub> O
8	Fe	Transition metal	FeCl <sub>2</sub> ·6H <sub>2</sub> O
9	Со	Transition metal	CoCl <sub>2</sub> ·6H <sub>2</sub> O
10	Ni	Transition metal	NiCl <sub>2</sub> ·6H <sub>2</sub> O
11	Cu	Transition metal	CuCl <sub>2</sub> ·2H <sub>2</sub> O
12	Zn	Transition metal	ZnSO <sub>4</sub> ·7H <sub>2</sub> O
13	Se	Diatomic nonmetal	Na <sub>2</sub> SeO <sub>3</sub>
14	Rb	Alkali metal	$RbCl_2$
15	Sr	Alkali earth metal	$Sr(NO_3)_2$
16	Y	Transition metal	Y(NO <sub>3</sub> ) <sub>3</sub> ·6H <sub>2</sub> O
17	Zr	Transition metal	$K_2ZrF_6$
18	Mo	Transition metal	Na <sub>2</sub> MoO <sub>4</sub> ·2H <sub>2</sub> O
19	Ag	Transition metal	AgNO <sub>3</sub>
20	Cd	Transition metal	$CdCl_2$
21	In	Post-transition metal	InCl <sub>2</sub> ·4H <sub>2</sub> O
22	Sn	Post-transition metal	SnCl <sub>2</sub> ·2H <sub>2</sub> O
23	Te	Metalloid	Na <sub>2</sub> TeO <sub>3</sub>
24	Ba	Alkali earth metal	Ba(CH <sub>3</sub> COO) <sub>2</sub>
25	La	Lanthanide	$La(NO_3)_3 \cdot 6H_2O$
26	Ce	Lanthanide	Ce(NO <sub>3</sub> ) <sub>3</sub> ·6H <sub>2</sub> O
27	Pr	Lanthanide	Pr(NO <sub>3</sub> ) <sub>3</sub> ·6H <sub>2</sub> O
28	Nd	Lanthanide	Nd(NO <sub>3</sub> ) <sub>3</sub> ·6H <sub>2</sub> O
29	Sm	Lanthanide	Sm(NO <sub>3</sub> ) <sub>3</sub> ·6H <sub>2</sub> O
30	Eu	Lanthanide	Eu(NO <sub>3</sub> ) <sub>3</sub> ·6H <sub>2</sub> O
31	Gd	Lanthanide	GdNO <sub>3</sub> ) <sub>3</sub> ·6H <sub>2</sub> O
32	W	Transition metal	Na <sub>2</sub> WO <sub>4</sub> ·6H <sub>2</sub> O
33	Au	Transition metal	HAuCl <sub>4</sub>
34	Hg	Transition metal	$HgCl_2$
35	Pb	Post-transition metal	$Pb(NO_3)_2$

Table S4. List of classified chemical types as the precursors for NM synthesis

## SI Appendix Text

**SI Appendix Text S1. Comparison of biosynthesis of NMs using bacteria and plant extracts.** The use of bacteria is better than using plant extract as the latter requires strong acids and heat for the preparation of the extract and additional reducing agents (i.e., sodium borohydride) for the supply of reducing power.

**SI Appendix Text S2. Previous study on biosynthesis of various NMs using recombinant** *E. coli* **co-expressing MT and PCS.** For the production of more various NMs by bacterial system, we previously developed a recombinant *E. coli* strain coexpressing MT and PCS, which allowed biosynthesis of noble NMs (Ag and Au), magnetic NMs (Fe, FeMn, FeCo, FeCoMn, FeCoNi and FeAg), quantum dots (QDs; ZnSe, CdZn, CdSe, CdSeZn, CdSeZnTe, CdTe, CdCs, PrGd, SrPr, SrGd, EuSe and AuCdSeZn) and metalloid NM (Te) upon addition of the corresponding precursors during the cultivation (8, 10, 11).

**SI Appendix Text S3. Expression of MT and PCS in** *E. coli* DH5α strain during the NMs synthesis. Expression of MT and PCS in *E. coli* was already investigated in the previous study (7). We further examined the changes of expression levels of the proteins over the time. MT is not visible due to its small size (~7.9 kDa) in the SDS-PAGE. The expression of PCS (~54.5 kDa) increased gradually up to 8 h after induction in the presence of Sn precursor, whereas it showed a slight decrease at 12 h after induction. In the culture supernatant, PCS was not observed in 16 h cultivation, suggesting that there was no leakage of PCS at least.

**SI Appendix Text S4.** *In vitro* **synthesis of single-element NMs.** Seven crystalline NMs synthesized *in vitro* were Mn<sub>3</sub>O<sub>4</sub>, Fe<sub>3</sub>O<sub>4</sub>, Cu<sub>2</sub>O, Ag, In(OH)<sub>3</sub>, SnO<sub>2</sub> and Au (Fig. 1*G*, *SI Appendix*, Figs. S7 and S8); the crystal structures of these NMs reconstructed based on the XRD data are shown in *SI Appendix*, Fig. S4. On the other hand, amorphous NMs were formed *in vitro* for Se, Zr, Te and Pb (Fig.1*H* and *SI Appendix*, Fig. S9). The other 23 elements, including Li, B, Al, V, Cr, Co, Ni, Zn, Rb, Sr, Y, Mo, Cd, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, W and Hg, resulted in no NM formation *in vitro*. Interestingly, seven lanthanide elements (La, Ce, Pr, Nd, Sm, Eu and Gd) tested all led to the formation of amorphous NMs *in vivo*, but no NM formation *in vitro*. *In vitro* synthesis of Mn, Ag and Sn NMs using the cell extract of the control *E. coli* strain harboring an empty vector resulted in crystalline Mn<sub>3</sub>O<sub>4</sub>, Ag and SnO<sub>2</sub> NMs, but again they were heterogeneous in size and shape (Fig. 1*I*), as observed above for *in vivo* biosynthesis.

**SI Appendix Text S5.** Pourbaix diagram analysis to predict NMs producibility. The Eh and pH values were measured during the *in vivo* syntheses of NMs for 12 h for each element, and the stability and predominant forms of elements were studied through Pourbaix diagram analyses (*SI Appendix*, Figs. S11-S13 and see *SI Appendix*, *Materials and Methods*). During the *in vivo* synthesis of NMs, Eh values decreased from +351.1~+356.4 mV to -182.2~-463.6 mV while the pH increased from 6.50 to 7.13~8.23. The results of Pourbaix diagram analyses were consistent with the *in vivo* experimental results; NM biosynthesis of correct product for 21 elements was confirmed (*SI Appendix*, Figs. S11 and S12).

SI Appendix Text S6. Pourbaix diagram analyses and measurements of the Eh and pH during *in vitro* biosynthesis. For *in vitro* synthesis of NMs, the Eh decreased from +349.4~+351.2 mV to -280.5~-445.2 mV and pH increased from 6.50 to 7.13~8.72 during the 12 h incubation (*SI Appendix*, Figs. S14-S16). As in the case of *in vivo* NM biosynthesis, the results of Pourbaix diagram analyses were consistent with the *in vitro* experimental results as well: among 34 elements examined, NM synthesis from 11 elements (*SI Appendix*, Figs. S14 and S15) and no NM synthesis from 14 elements are recapitulated in the Pourbaix diagram analysis (*SI Appendix*, Fig. S16). However, no NM synthesis from nine elements (B, Al, V, Cr, Y, Mo, Sm, Eu and Gd) are unexplainable using Pourbaix diagram.

SI Appendix Text S7. Two separate single-element crystalline NMs synthesized *in vivo* at pH 7.5. In the case of Co and Ce, two separate single-element crystalline NMs were synthesized *in vivo* at pH 7.5:  $\beta$ -Co(OH)<sub>2</sub> and Co<sub>3</sub>O<sub>4</sub>; Ce(OH)<sub>3</sub> and CeO<sub>2</sub> (*SI Appendix*, Fig. S19*B*). For the former case, it has been reported that chemical synthesis of Co NM resulted in the formation of thermodynamically less stable  $\alpha$ -Co(OH)<sub>2</sub>, which is rapidly converted to  $\beta$ -Co(OH)<sub>2</sub> (12). Then,  $\beta$ -Co(OH)<sub>2</sub> is gradually transformed into Co<sub>3</sub>O<sub>4</sub>, resulting in the detection of only  $\beta$ -Co(OH)<sub>2</sub> and Co<sub>3</sub>O<sub>4</sub>, but no  $\alpha$ -Co(OH)<sub>2</sub> by XRD analysis. For the latter case, Pourbaix diagram analysis indicates that the *in vivo* biosynthesis condition is initially favorable for the formation of CeO<sub>1</sub> (*SI Appendix*, Fig. S17).

SI Appendix Text S8. Formation of crystalline NMs or amorphous NMs, or no NM formation in vitro at pH 7.5. Li, Rb, Sr and W (SI Appendix, Fig. S16) were excluded due to the same reason described for *in vivo* reactions. Pourbaix diagrams of 23 elements, that did not form crystalline NMs at pH 6.5, were analyzed and the Eh and pH of *in vitro* reactions were mapped (SI Appendix, Figs. S22 and S23). By raising the pH of in vitro reactions to 7.5 for 12 h, 11 crystalline NMs were synthesized as their predicted crystalline phases:  $\beta$ -Co(OH)<sub>2</sub>,  $\beta$ -Ni(OH)<sub>2</sub>, ZnO,  $\beta$ -Cd(OH)<sub>2</sub>, La(OH)<sub>3</sub>, CeO<sub>2</sub>, Pr(OH)<sub>3</sub>, Nd(OH)<sub>3</sub>, Sm(OH)<sub>3</sub>, Eu(OH)<sub>3</sub> and Gd(OH)<sub>3</sub> (Fig. 2D, SI Appendix, Figs. S25 and S27); the crystal structures of these NMs reconstructed based on the XRD data are shown in *SI Appendix*, Fig. S27. It should be noted that abrupt change of the initial pH to 7.5 caused appearance of some precipitates (i.e., Ni, Zn, Nd, and Sm) in 30 min (SI Appendix, Fig. S24), which seemed to grow to crystalline NMs after prolonged reaction for 12 h. In the case of Pb, the Pourbaix diagram predicted formation of Pb(OH)<sub>2</sub>, but Pb<sub>3</sub>(NO)<sub>3</sub>(OH)<sub>5</sub> and Pb<sub>2</sub>(NO)<sub>3</sub>(OH)<sub>3</sub> NMs were synthesized *in vitro*. For Se, Y, Zr and Te, amorphous NMs were synthesized by in vitro reaction at pH 7.5 (Fig. 2E and SI Appendix, Fig. 28). In the cases of B, Al, V, Cr, Mo and Ba, NMs were not synthesized by *in vitro* reactions even at pH 7.5 (SI Appendix, Fig. S23B).

SI Appendix Text S9. Amorphous NMs synthesized *in vivo* and *in vitro* at pH 7.5. It was previously reported that biosynthesized Se NM is typically amorphous under ambient conditions (10). In the case of Y, Y(OH)<sub>3</sub> crystalline NM could be obtained by increasing the initial pH higher than 13 under hydrothermal precipitation methods (11). However, amorphous Y NM was synthesized *in vivo* and *in vitro* at initial pH 7.5 (Fig. 2*C*, *E*). For

Mo and Ba, the Pourbaix diagram predicted biosynthesis of crystalline MoO<sub>2</sub> and BaCO<sub>3</sub> by both *in vivo* and *in vitro* reactions, but only *in vivo* reaction resulted in their biosynthesis in our experiments (*SI Appendix*, Fig. S23*B*). For these several elements, further studies are needed to better understand the mechanisms underlying the formation of NMs with respect to crystallinity and producibility.

SI Appendix Text S10. *a*-Fe<sub>2</sub>O<sub>3</sub> NM synthesized *in vivo* and *in vitro* by 6-hour reaction. Biosynthesis of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM was accompanied with generation of two byproducts  $\beta$ -FeOOH and  $\alpha$ -FeOOH (*SI Appendix*, Figs. S30*A*-*D* and S31*A*-*F*). To obtain only  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NM without the byproducts, the mixture of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>,  $\beta$ -FeOOH and  $\alpha$ -FeOOH was calcinated at 800°C in the presence of air for 2 h. The resulting  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> NMs synthesized *in vivo* and *in vitro* showed different sizes of 12.27 ± 3.37 nm and 14.68 ± 4.97 nm, respectively; the crystalline structure was reconstructed based on the XRD data (*SI Appendix*, Figs. S30*E*-*H* and S31*G*-*K*).

SI Appendix Text S11. Formation of single crystalline NMs or amorphous NMs, or no NM formation *in vivo* for those elements that were predicted to form crystalline multi-element NMs. Although the Pourbiax diagram analysis predicted possibility of synthesizing nine crystalline NMs for Ag<sub>3</sub>H<sub>2</sub>VO<sub>5</sub>, Ag<sub>2</sub>Se, Pb<sub>2</sub>V<sub>2</sub>O<sub>7</sub>, Cu<sub>2</sub>Se, SrSeO<sub>3</sub>, Al<sub>4</sub>(OH)<sub>10</sub>SO<sub>4</sub>, ZnCr<sub>2</sub>O<sub>4</sub>, CoS and MoS<sub>2</sub> by *in vivo* biosynthesis (*SI Appendix*, Fig. S32), single crystalline Ag NM was synthesized for V/Ag and Se/Ag combinations (*SI Appendix*, Fig. S37*A*), single amorphous Pb NM was synthesized for V/Pb combination, and single amorphous Se NM was synthesized for Cu/Se and Se/Sr combinations (*SI Appendix*, Fig. S37*B*). The other four bi-elemental combinations (Al/S, S/Co, S/Mo and Cr/Zn) resulted in no *in vivo* NM biosynthesis.

**SI Appendix Text S12. Fourty-two bi-elemental combinations to biosynthesize multielement crystalline NMs.** We examined producibility of the folloing 42 bi-elemental combinations: magnetic NMs including Mn/Ni, Mn/Zn and Co/Ni; heavy metal free QDs including S/Mn, S/In, S/Sn, S/W, Mn/Te, Se/Mo, Ag/Te, In/Te, Sn/Te and Te/Hg; and various randomly selected NMs including Li/B, Li/W, S/V, V/Se, V/Sn, V/Pb, Cr/Ag, Cu/La, Cu/Ce, Cu/Pr, Cu/Nd, Cu/Sm, Cu/Eu, Cu/Gd, Se/W, Rb/La, Sr/Ag, Y/Cd , Zr/W, Ag/Sn, Cd/La, Cd/Ce, Cd/Pr, Cd/Nd, Cd/W, Sn/W, Te/Au, La/Pb and W/Au (*SI Appendix*, Fig. S34)

SI Appendix Text S13. Formation of single crystalline NMs or amorphous NMs, or no NM formation *in vivo* for those elements that were predicted not to form multielement NMs. For the five bi-elemental combinations, including Mn/Te, Ag/Sn, In/Te, Sn/Te and La/Pb, two separate NMs (e.g., Mn NM and Te NM for Mn/Te) were biosynthesized (*SI Appendix*, Fig. S37*C*). Six combinations including Li/B, Li/W, S/V, S/W, Co/Ni and Cd/W produced no *in vivo* NM synthesis. The other 28 bi-elemental combinations resulted in biosynthesis of NMs of only one element (*SI Appendix*, Figs. S34*C* and S37*D*). In single element experiments shown above, lanthanides including Y, La, Ce, Pr, Gd, Nd, Sm and Eu formed amorphous NMs. Bi-element combinations including these lanthanides resulted in formation of single lanthanide amorphous NMs. Two elements, V and Rb, which did not form NMs in bi-element experiments as observed in single-element experiments. Although Se and Pb formed amorphous NMs while Cd and W did not form NMs in single-element experiments, they formed multi-element crystalline NMs as in Ag<sub>2</sub>WO<sub>4</sub>, CdSe, PbMoO<sub>4</sub> and PbWO<sub>4</sub>. When bi-element NM biosynthesis involving Cu and lanthanides such as Cu/La, Cu/Ce, Cu/Pr, Cu/Gd, Cu/Nd, Cu/Sm and Cu/Eu was performed, it was interesting to observe that single amorphous lanthanide NMs without crystalline Cu<sub>2</sub>O NM were formed. Further studies are needed to understand the mechanisms underlying these phenomena.

SI Appendix Text S14. Formation of single crystalline or amorphous NMs, two separate single-element NMs, or no NM synthesized *in vitro*. Single crystalline Ag NM was biosynthesized for V/Ag and Se/Ag combinations (*SI Appendix*, Fig. S40A), only single amorphous Pb NM for V/Pb combination, and single amorphous Se NM for Cu/Se and Se/Sr combinations (*SI Appendix*, Fig. S40B); these results are the same as those observed in *in vivo* experiments. In the case of Ag/Sn combination, two separate singleelement NMs were *in vitro* synthesized as observed in *in vivo* experiment (*SI Appendix*, Fig. S40C). In the cases of combinations including Te element (i.e. Mn/Te, In/Te and Sn/Te), single crystalline Mn<sub>3</sub>O<sub>4</sub>, In(OH)<sub>3</sub> and SnO<sub>2</sub> NMs without single Te NM formation (*SI Appendix*, Fig. S40D), which are different from *in vivo* biosynthesis results. The other 19 combinations (i.e. S/V, S/Mo, Co/Ni, Rb/La, Cd/Y, Cd/La, Cd/Ce, Cd/Pr, Cd/Nd and La/Pb) is consistent with that observed in *in vitro* single elemental screening results. Differently from *in vivo* biosynthesis experiments, no NM was *in vitro* synthesized for bielemental combinations of Cu and lanthanides, while Cu<sub>2</sub>O crystalline NM was *in vitro* biosynthesized when only Cu was present.

SI Appendix Text S15. Multi-element NMs synthesized in vivo and in vitro at pH 7.5. In the four case of S/Co, Se/Ag, Cu/Se and Se/Sr bi-elemental combinations, single crystalline NMs, including CoO(OH) and Ag (SI Appendix, Figs. S41A and S43A) and single amorphous Se NM were biosynthesized (SI Appendix, Fig. S43B). For Ag/Sn and La/Pb, two individual NMs were in vitro synthesized in both cases (SI Appendix, Fig. S42C). For most of the bi-elemental combinations tested *in vivo* and *in vitro*, single NMs were biosynthesized even at pH 7.5 (SI Appendix, Figs. S41C and S42D). For V/Ag, crystalline NaVO<sub>2</sub>, NaAg<sub>3</sub>O<sub>2</sub> and Ag<sub>2</sub>O NMs were biosynthesized both *in vivo* and *in vitro* at pH 7.5 (SI Appendix, Fig. S43A-E), differently from formation of single crystalline Ag NM at pH 6.5 (SI Appendix, Figs. S37A and S40A). For Cr/Zn, crystalline Na<sub>4</sub>CrO<sub>4</sub> and  $Zn(OH)_2$  NMs were produced at pH 7.5 (SI Appendix, Fig. S43F-J) while no NM biosynthesis was observed at pH 6.5. For V/Pb combination, crystalline Pb<sub>5</sub>(VO<sub>4</sub>)<sub>3</sub>OH NM was biosynthesized at pH 7.5, although solid phase  $Pb_3(VO_4)_2$  or  $Pb_2V_2O_7$  were predicted by Pourbaix diagram analysis (SI Appendix, Figs. S36D, S36E and S44). No NM was biosynthesized for Al/S and S/Mo at pH 7.5, although the Pourbaix diagram predicted formation of Al<sub>4</sub>(OH)<sub>10</sub>SO<sub>4</sub> and MoS<sub>2</sub> (*SI Appendix*, Fig. S32).

**SI Appendix Text S16. Magnetic properties of the crystalline NMs synthesized** *in vivo* **after calcination.** For *in vivo* biosynthesized NMs, the magnetic properties were measured with the centrifuged samples containing NMs as well as cell debris co-precipitated. When

using the *in vitro* method, the supernatant of centrifuged cell lysate was used for NM biosynthesis, and thus no cell debris was present. After cell debris of *in vivo* synthesized magnetic NMs were removed by calciation at 800 °C, the NMs showed paramagnetic (Mn<sub>3</sub>O<sub>4</sub> and ZnMn<sub>2</sub>O<sub>4</sub>), ferromagnetic (Fe<sub>3</sub>O<sub>4</sub> and CoFe<sub>2</sub>O<sub>4</sub>) and superparamagnetic (NiFe<sub>2</sub>O<sub>4</sub> and ZnFe<sub>2</sub>O<sub>4</sub>) properties (black lines in *SI Appendix*, Fig. S54).

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