Electronic Supplementary Information

Effect of Ag evolution process on ordering transition for $L1_0$-FePt nanoparticles synthesized by Ag addition

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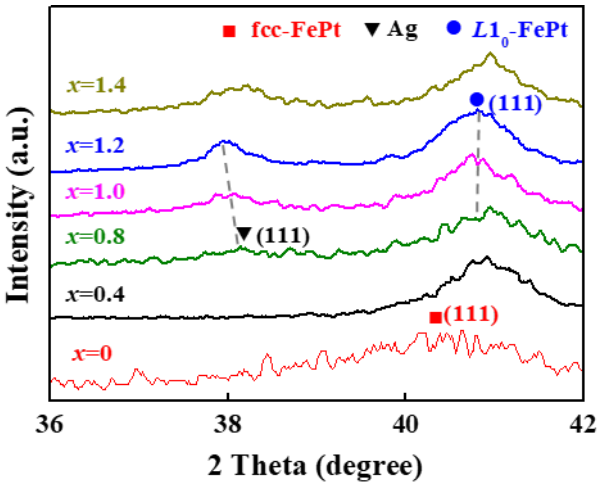

Fig. S1. Enlarged sections of the XRD patterns of the as-synthesized NPs with various Ag/Pt molar ratios (*x*).

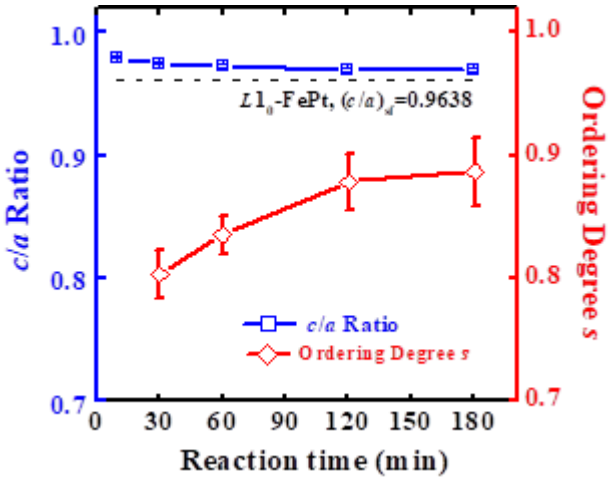

Fig. S2. The *c*/*a* ratios and ordering degrees *s* of the NPs collected at 360 °C for different reaction times.

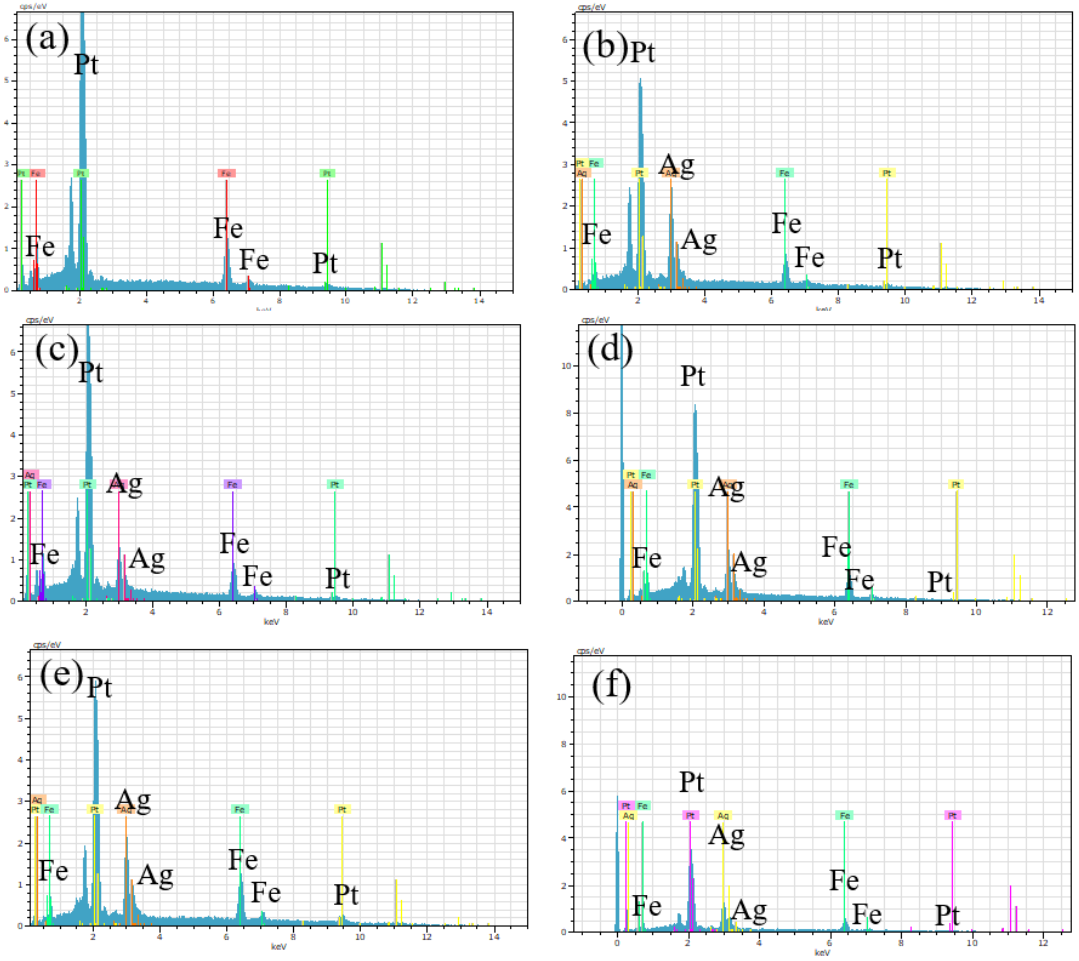

Fig. S3. EDS spectrums of NPs synthesized with various Ag/Pt mole ratios (*x*): (a) *x*=0, (b) *x*=0.4, (c) *x*=0.8, (d) *x*=1.0, (e) *x*=1.2, (f) *x*=1.4.

Computational details

The Vienna Ab-initio Simulation Package (VASP) software package developed by the University of Vienna was used to perform first-principles calculations based on spin polarization density functional theory (DFT).[1] The projector-augmented wave (PAW) pseudopotentials were used for electron-ion interactions.[2] At the equiatomic composition, $L1_0$-FePt is comprised of alternating atomic planes of Fe and Pt along the unit-cell *c* axis. The 2 × 2 × 2 supercell model with 32 atoms was used to calculate the structure of $L1_0$-FePt. The size of the supercell has been fully verified.[3] The disordered model of the FePt binary alloy was generated by the gensqs and mcsqs codes in the Alloy Theory Automation Toolkit (ATAT).[4-6] The correlation functions of two clusters and three clusters match perfectly with the values of the ideal random structure. The models of the ordered and disordered are shown in Fig. S4.

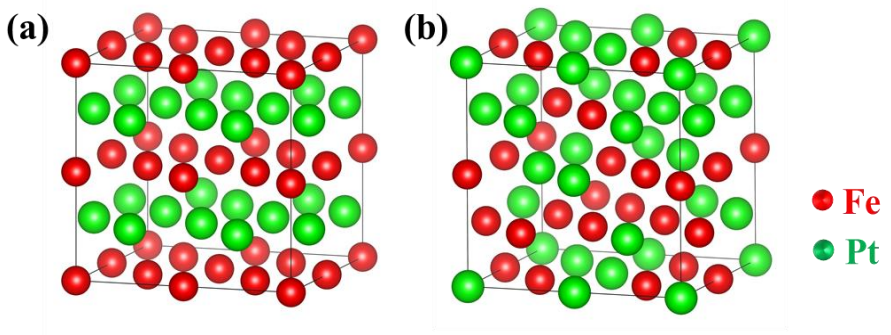

Fig. S4. (a) ordered $L1_0$-FePt supercell structure, (b) disordered fcc FePt supercell structure.

The substitution formation energies (E_{sub}) is the energy required for the third element to replace the main phase element, which can be used to measure the difficulty of the third element doping in the main phase the E_{sub} can be obtained using the following equation[7]:

$$E_{sub} = E_{doped} + E_{pure} + \mu_{lose} - \mu_{x} \quad \text{(S1)}$$

where the E_{doped} and E_{pure} are the total energies of FeAgPt and FePt systems respectively. μ_{lose} is the chemical potential of the substituted atoms, μ_{x} is the chemical potential of the Ag elements. All μ_{i} are defined as the value referred to as its most stable phase.

The vacancy formation energy (E_{vac}) is used to describe the energy required to form a point defect. The calculation formula of E_{vac} is as follows:

$$E_{vac} = E_{N-1} - E_{N} + \mu_{x} \quad \text{(S2)}$$

where E_{N-1} and E_{N} are the total energy of the vacancy system and the perfect system, respectively, and μ_{x} is the chemical potential of the atom at the vacancy.

The results show that the E_{sub} of the Ag atom replacing Fe site (28.8 meV·atom^{-1}) are lower than that of replacing Pt site (40.1 meV·atom^{-1}), indicating that Fe atoms were easier to be replaced by Ag-elements than Pt atoms. The vacancy formation energies E_{vac} of Fe atom decreased from 48.0 to 43.8 meV·atom^{-1}, and E_{vac} of Pt atom decreased from 72.4 to 48.5 meV·atom^{-1}.

References

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