Electronic Supplementary Information (ESI)

Preparation of solid-solution-type Fe–Co nanoalloys by synchronous deposition of Fe and Co using dual arc plasma guns

Masaaki Sadakiyo,[a,b] Minako Heima,[a,b] Tomokazu Yamamoto,[c] Syo Matsumura,[c] Masashi Matsuura,[d] Satoshi Sugimoto,[d] Kenichi Kato,[b,e] Masaki Takata,[e] and Miho Yamauchi*[a,b,f]

[a] *International Institute for Carbon-Neutral Energy Research (WPI-I2CNER), Kyushu University, 744 Moto-oka, Nishi-ku, Fukuoka 819-0395, Japan.* [b] *Core Research for Evolutional Science and Technology (CREST), Japan Science and Technology Agency (JST), 7 Goban-cho, Chiyoda-ku, Tokyo 102-0076, Japan.* [c] *Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University, 744 Moto-oka, Nishi-ku, Fukuoka 819-0395, Japan.* [d] *Department of Materials Science, Graduate School of Engineering, Tohoku University, 6-6-02 Aramaki Aza Aoba, Aoba-ku, Sendai 980-8579, Japan.* [e] *RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan.* [f] *Department of Chemistry, Faculty of Science, Kyushu University, 6-10-1 Hakozaki, Higashi-ku, Fukuoka 812-8581, Japan*

E-mail: yamauchi@i2cner.kyushu-u.ac.jp

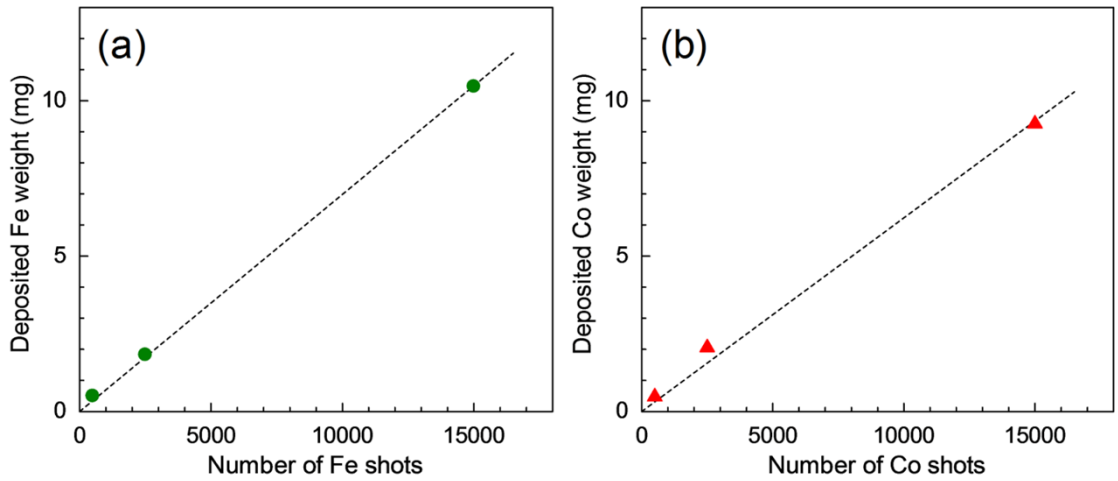

Fig. S1 Deposited metal weight vs. number of shots of (a) Fe and (b) Co sources, estimated by ICP-AES measurements.

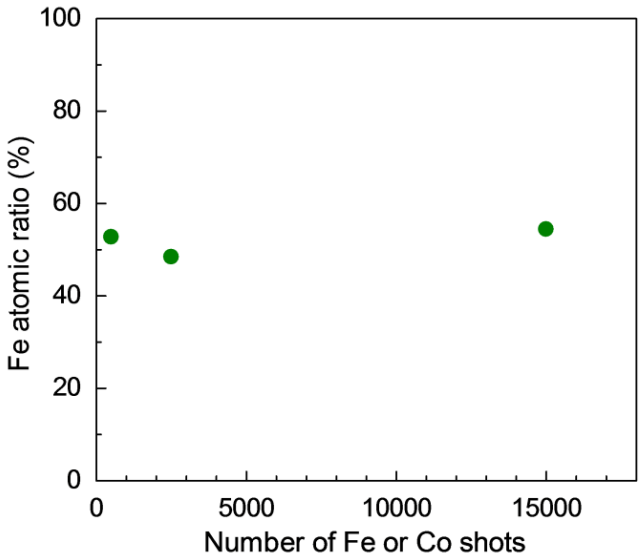

Fig. S2 Fe atomic ratio vs. number of shots of Fe (or Co), determined by ICP-AES measurements.

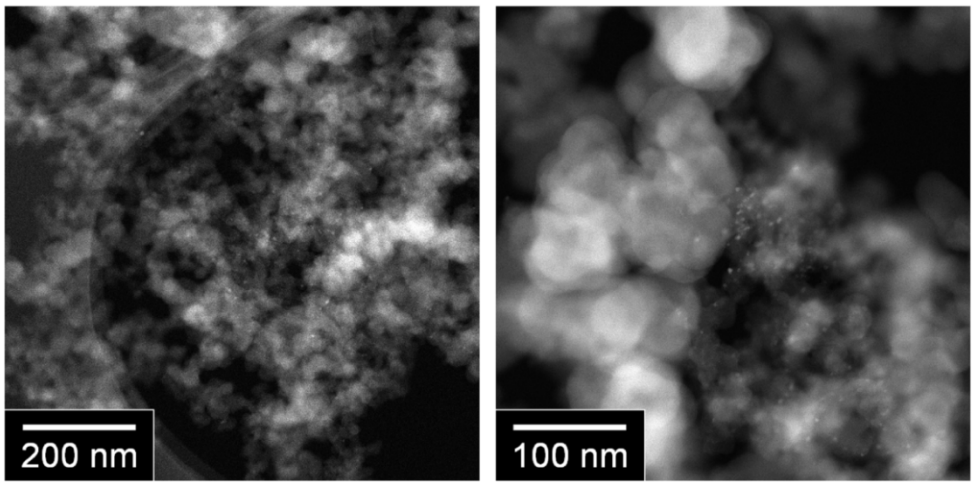

Fig. S3 Low magnification HAADF-STEM images of **FeCo^{5000}**. Small white dots correspond to the deposited FeCo particles. There also exist bare surface of carbon supports.

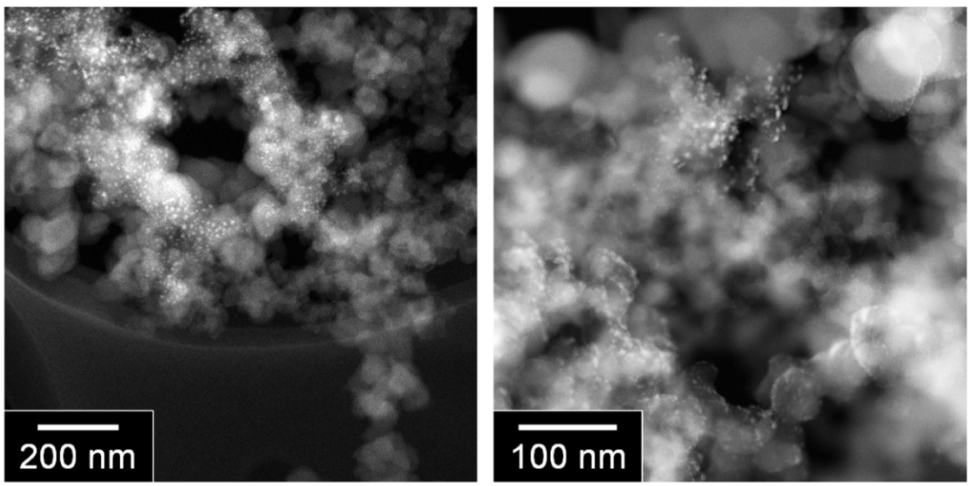

Fig. S4 Low magnification HAADF-STEM images of **FeCo^{3000}**. Small white dots correspond to the deposited FeCo particles. There also exist bare surface of carbon supports.

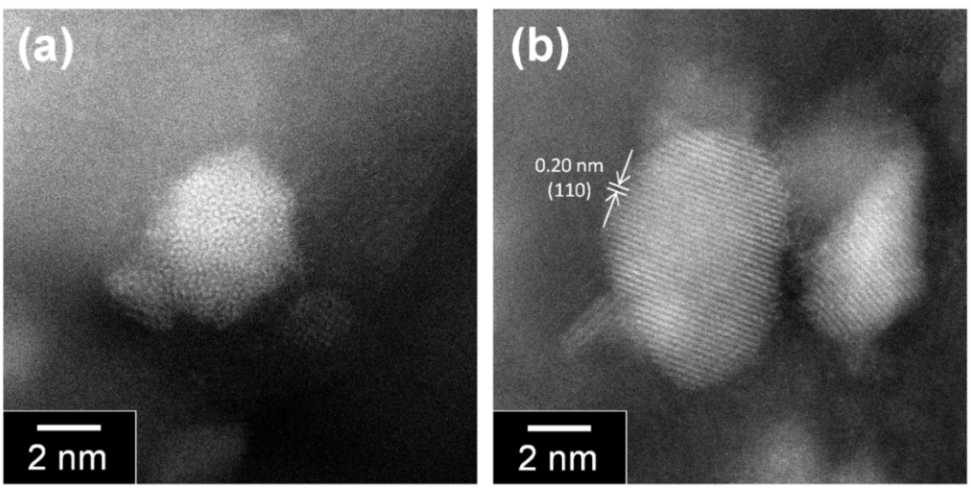

Fig. S5 HR–STEM images of (a) **FeCo^{5000}** and (b) **FeCo^{30000}**.

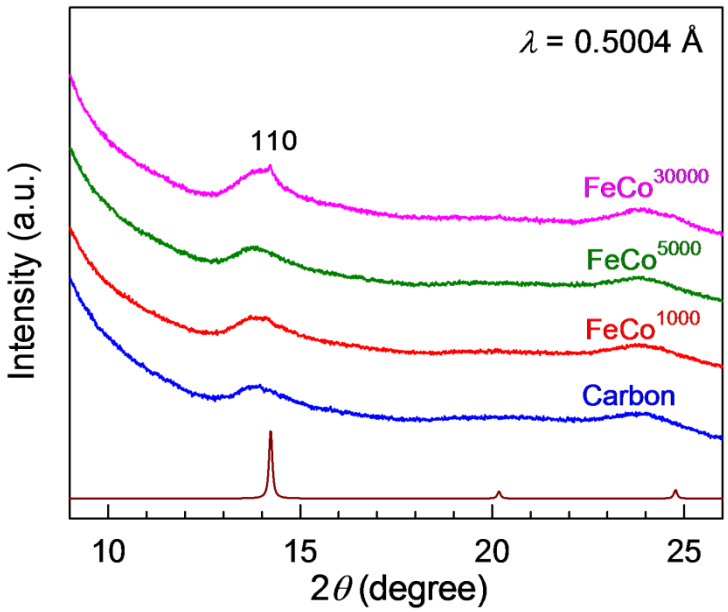

Fig. S6 XRPD patterns of a simulation of B2-structured Fe–Co alloy (brown), carbon support (blue), **FeCo^{1000}** (red), **FeCo^{5000}** (green), and **FeCo^{30000}** (pink).

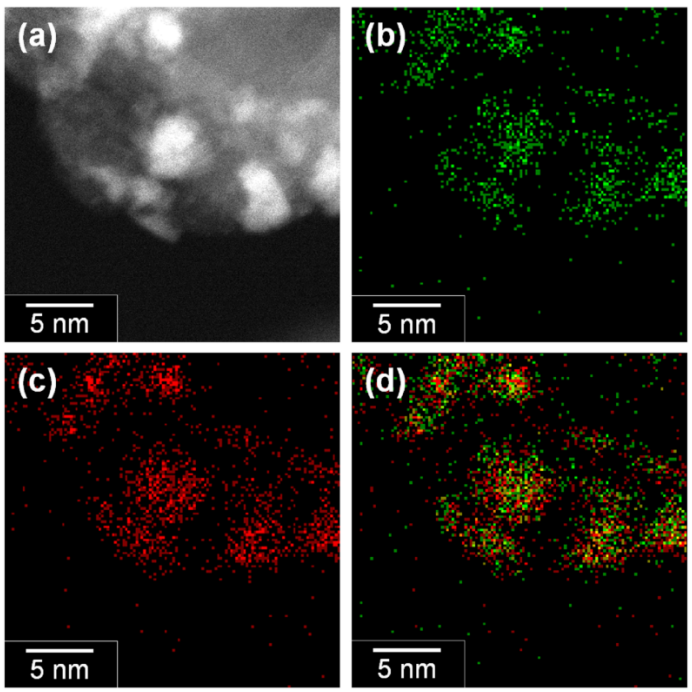

Fig. S7 (a) HAADF–STEM image of grains of **FeCo$_{5000}$**. STEM–EDX map of (b) Fe-K and (c) Co-K signals. (d) overlap image of Fe-K and Co-K signals. Green and red correspond to Fe-K and Co-K signals, respectively.

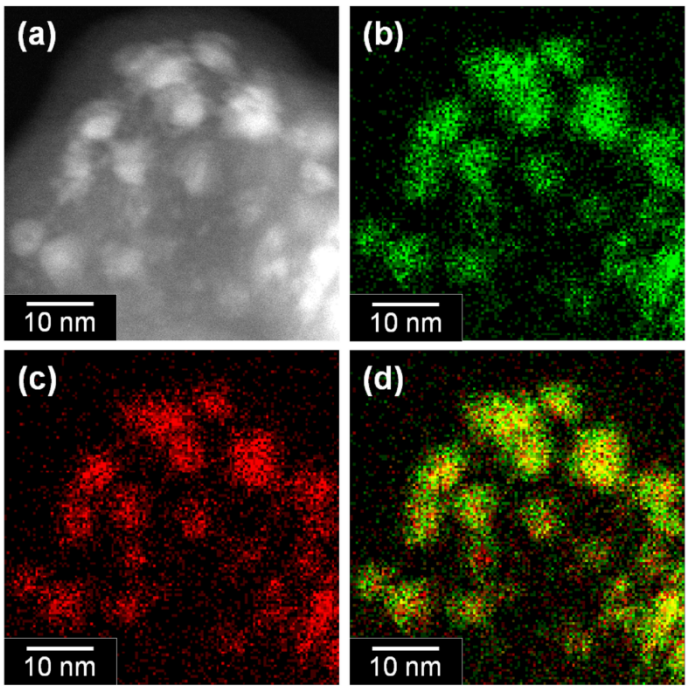

Fig. S8 (a) HAADF–STEM image of grains of **FeCo^{30000}**. STEM–EDX map of (b) Fe-K and (c) Co-K signals. (d) overlap image of Fe-K and Co-K signals. Green and red correspond to Fe-K and Co-K signals, respectively.

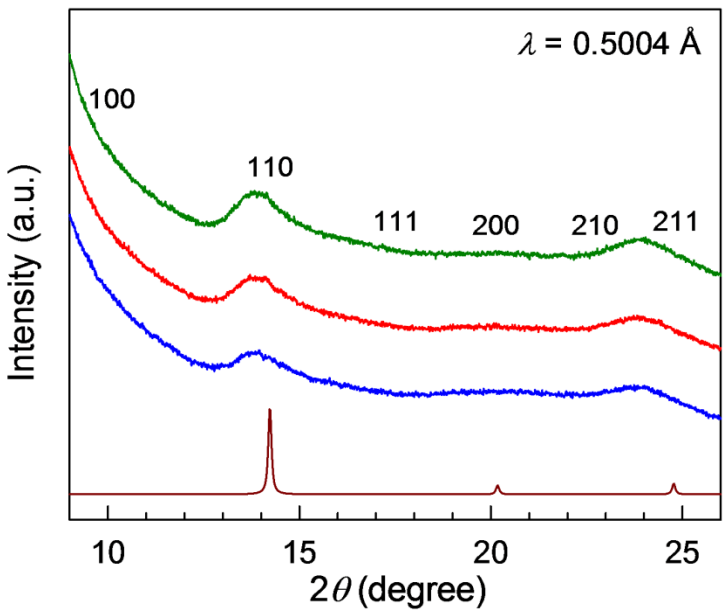

Fig. S9 XRPD patterns of a simulation of B2-structured Fe-Co alloy (brown), carbon support (blue), as-prepared **FeCo^{1000}** (red), and **FeCo^{1000}** treated at 800 °C under 5% H_2/Ar gas flow (green).

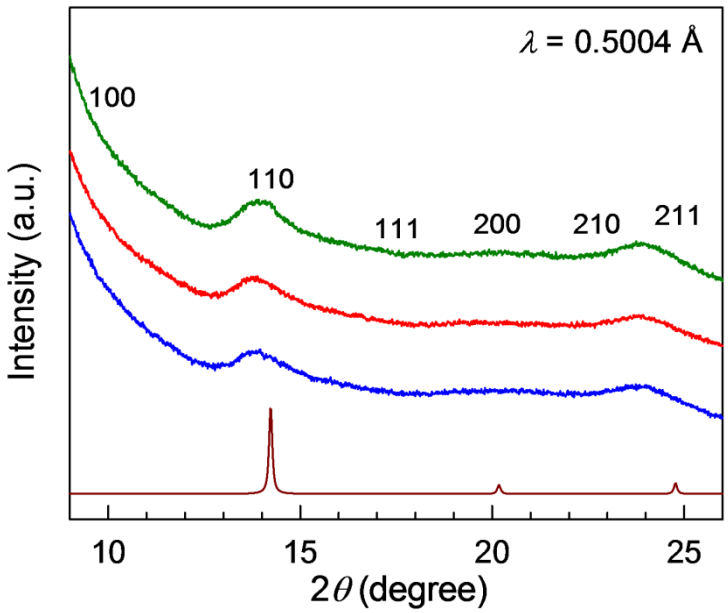

Fig. S10 XRPD patterns of a simulation of B2-structured Fe-Co alloy (brown), carbon support (blue), as-prepared **FeCo^{5000}** (red), and **FeCo^{5000}** treated at 800 °C under 5% H_2/Ar gas flow (green).

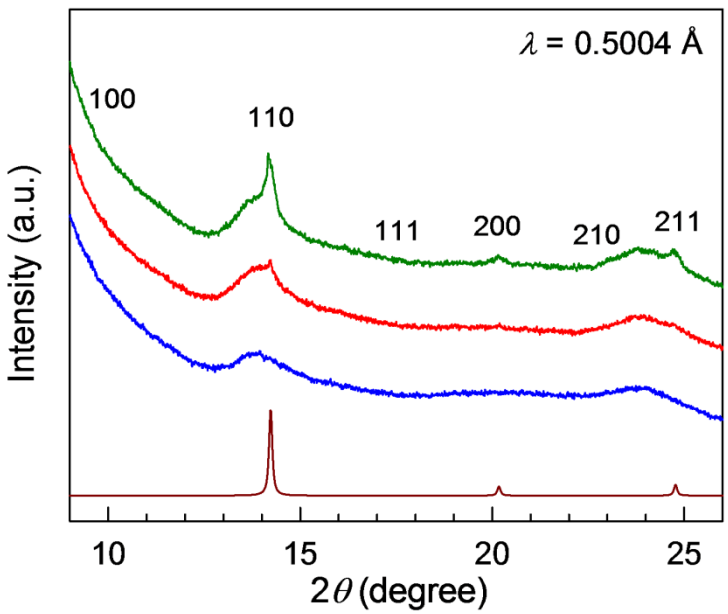

Fig. S11 XRPD patterns of a simulation of B2-structured Fe–Co alloy (brown), carbon support (blue), as-prepared **FeCo^{30000}** (red), and **FeCo^{30000}** treated at 800 °C under 5% H_2/Ar gas flow (green).

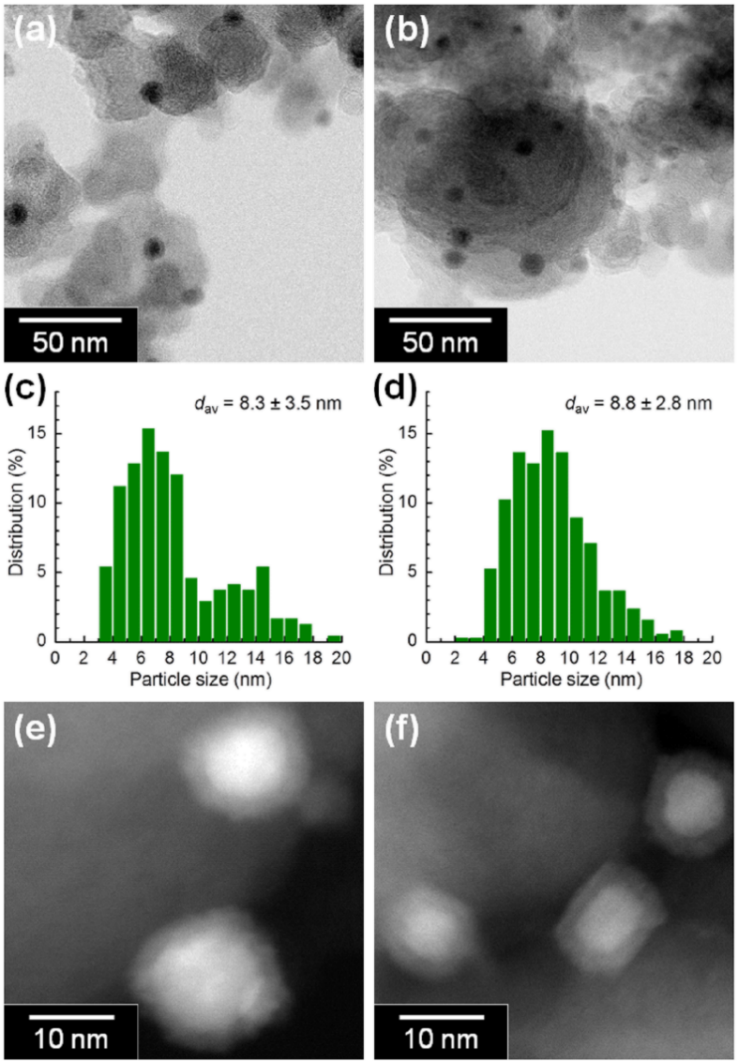

Fig. S12 STEM images of (a) **FeCo^{5000}** and (b) **FeCo^{30000}** after hydrogen treatment at 800 °C under H_2/Ar mixed gas flow. Particle size distributions of (c) **FeCo^{5000}** and (d) **FeCo^{30000}**. HAADF–STEM images of (e) **FeCo^{5000}** and (f) **FeCo^{30000}** after the hydrogen treatment.

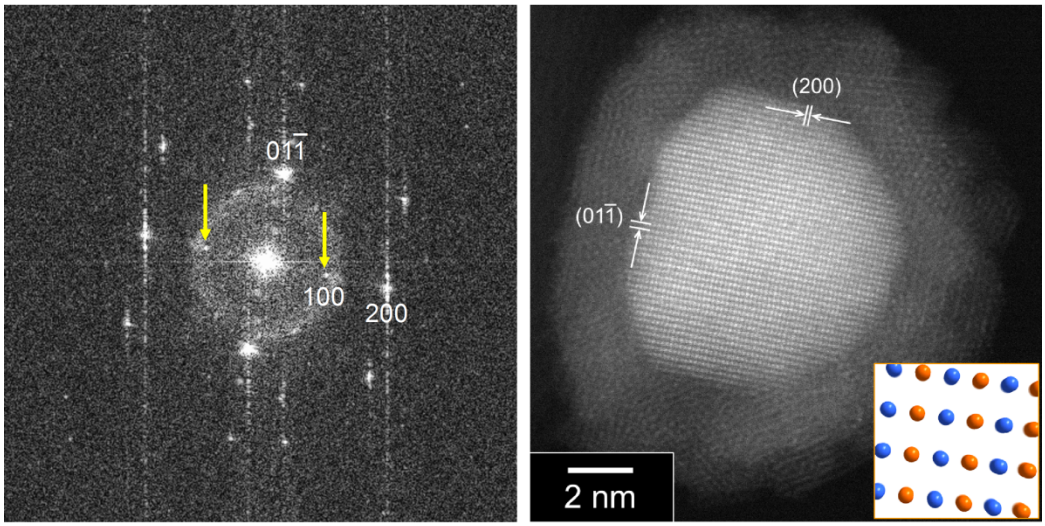

Fig. S13 (left) A magnified FFT image of (right) the HR-STEM image of **FeCo^{5000}**. The yellow arrows indicate the characteristic spots of ordered B2 structure (e.g. (100) plane).

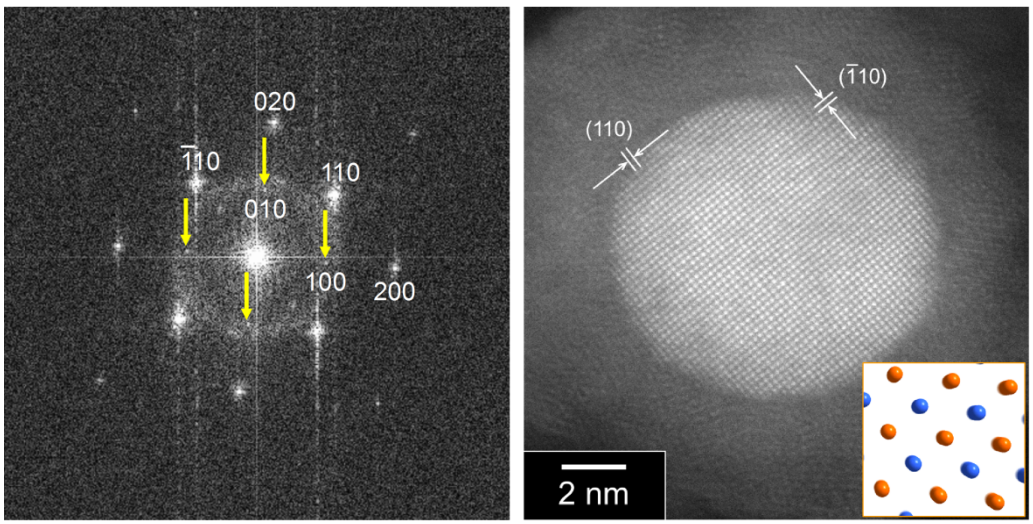

Fig. S14 (left) A magnified FFT image of (right) the HR-STEM image of **FeCo^{30000}**. The yellow arrows indicate the characteristic spots of ordered B2 structure (e.g. (100) plane).

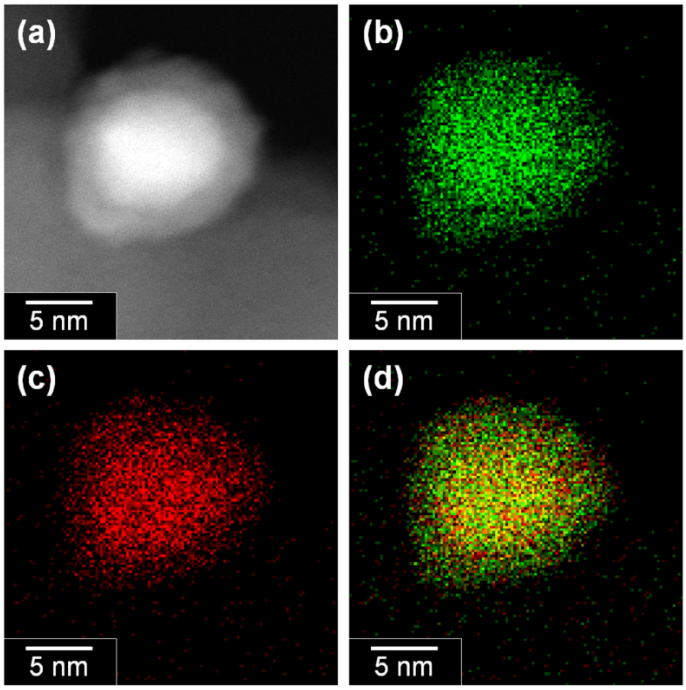

Fig. S15 (a) HAADF–STEM image of a nanoparticle of **FeCo^{5000}** after hydrogen treatment at 800 °C under H_2/Ar mixed gas flow. STEM–EDX map of (b) Fe-K and (c) Co-K signals. (d) overlap image of Fe-K and Co-K signals. Green and red correspond to Fe-K and Co-K signals, respectively.

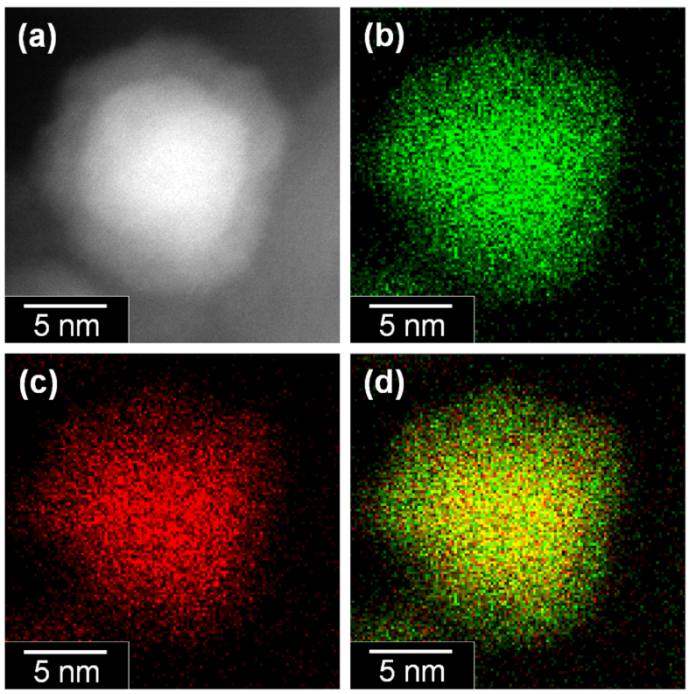

Fig. S16 (a) HAADF–STEM image of a nanoparticle of **FeCo^{30000}** after hydrogen treatment at 800 °C under H_2/Ar mixed gas flow. STEM–EDX map of (b) Fe-K and (c) Co-K signals. (d) overlap image of Fe-K and Co-K signals. Green and red correspond to Fe-K and Co-K signals, respectively.

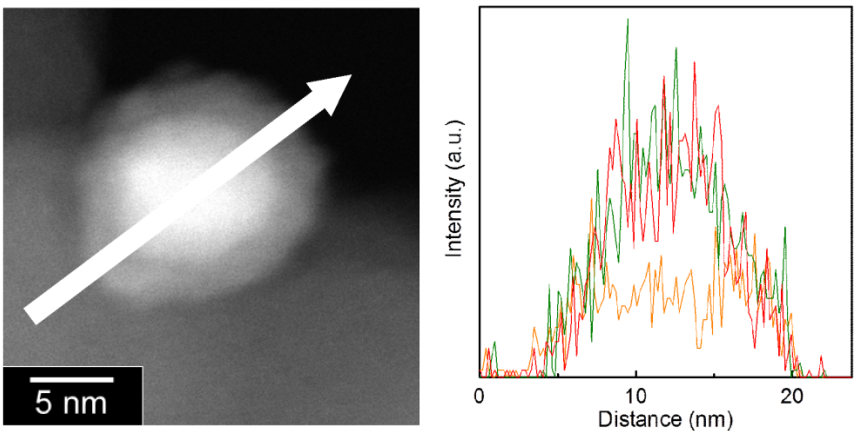

Fig. S17 A line profile of EDX signals of Fe-K, Co-K, and O-K peaks of **FeCo^{5000}** after hydrogen treatment at 800 °C under H_2/Ar mixed gas flow. Green, red, and orange correspond to Fe-K, Co-K, and O-K signals, respectively.

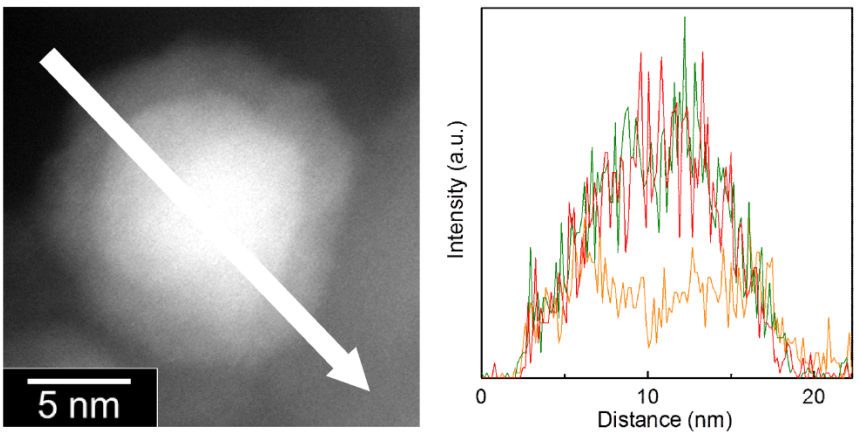

Fig. S18 A line profile of EDX signals of Fe-K, Co-K, and O-K peaks of **FeCo^{30000}** after hydrogen treatment at 800 °C under H_2/Ar mixed gas flow. Green, red, and orange correspond to Fe-K, Co-K, and O-K signals, respectively.