Supporting Information

Metal-Redox for MnAl-Based Ternary Magnetic Nanocrystals

Jian Shen, Jiang Li and Shenqiang Ren*

Department of Mechanical Engineering, Temple University, Philadelphia, Pennsylvania 19122, United States

E-mail: <u>shenqiang.ren@temple.edu</u>



Figure S1. (a) Magnetization saturation and coercivity depends on the AI precursors, (b) Magnetization saturation and coercivity depends on the injection atom ratio of Mn and AI, (c) Magnetization saturation and coercivity depends on the reaction temperature, (d) Magnetization saturation and coercivity depends on the AI precursor injection temperature.

The challenges of MnAl nanoalloy synthesis route in this study are controlling the ratio of Mn and Al, the formation speed of Al and Mn nanoparticles and the formation of MnAl alloy. Therefore, the aluminum precursors, the injection atom ratio of Mn and Al, reaction temperature and the Al precursor injection temperature are key factors. Figures S1 shows effects of key factors on magnetization saturation and coercivity of MnAl alloy. As shown in Fig. S1, AlCl₃ is a good precursor for MnAl alloy synthesis. At the same time, Mn: Al 4:1, reaction temperature (513 K) and the Al precursors injection temperature (513 K) make the MnAl alloy with relative high magnetic performance.



Figure S2. (a) Magnetic-hysteresis loops of CoAl and CoMn, (b) TEM, STEM, Al elemental mapping images in red and Co elemental mapping images in yellow of CoAl (from left to right), (c) TEM, STEM, Mn elemental mapping images in red and Co elemental mapping images in yellow of MnAl (from left to right).

In order to eliminate contribution of CoMn and CoAl to the magnetic properties of ternary CoMnAl nanoalloy, we synthesized CoMn and CoAl as control samples. Figure S2a shows magnetic-hysteresis loops of CoAl and CoMn. The magnetization saturation and coercivity of CoMn is 0.81 emu/g and 99 Oe, respectively. The corresponding value of CoAl is 0.82 emu/g and 67 Oe, respectively. The magnetic

properties indicate that the contribution of CoMn and CoAl to the ternary CoMnAl nanoalloy can be neglected. Figure S2b and S2c show TEM, STEM images and EDX mapping of corresponding elements. For AlCo, it obviously shows that Al and Co phase separate. For CoMn, the stoichiometry of MnCo is 60:40, which displays antiferromagnetic properties.



Figure S3. Magnetic–hysteresis loops of ternary CoMnAl with Mn90.8Al0.1Co9.1under different annealing temperature.

To increase crystalline and reduce the metal oxide in the surface of the alloy, the ternary CoMnAl with Mn_{90.8}Al_{0.1}Co_{9.1} was annealed at the different temperature. Figure S3 shows that the optimal magnetic performance can be obtained after annealing at 823K. Lower or higher than 823 K, the Ms and coercivity simultaneously decrease. Therefore, in this study, we select 823K as annealing temperature.



Figure S4. (a) Effect of the injection atom ratio of Mn and Al on magnetic-hysteresis loops of ternary CoMnAl alloy with 5% Co injection atomic percentage under the reaction temperature 513 K with 1 hour, (b) Effect of the Co injection atomic percentage of on magnetic-hysteresis loops of ternary CoMnAl alloy with Mn:Al 5:1 under the reaction temperature 513 K with 1 hour.

Reaction temperature and time affect the stoichiometry of aluminum, cobalt and manganese, which result in the difference in magnetic performance. In order to optimize the stoichiometry of Al, Co and Mn, the injection atom ratio of Mn and Al and the injection atomic percentage of Co are tuned.



Figure S5. Intergrated EDX spectrum images of $Mn_{90.8}Al_{0.1}Co_{9.1}$, $Mn_{90.1}Al_{0.9}Co_{9.0}$, $Mn_{68.2}Al_{17.9}Co_{13.9}$, $Mn_{84.2}Al_{8.1}Co_{7.6}$, $Mn_{40.5}Al_{55.4}Co_{4.1}$ (from top to bottom)