$D0_3$-ordered Fe_3Al nanopowders synthesised by low oxygen induction thermal plasma

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Abstract: With a low oxygen induction thermal plasma system, $D0_3$-ordered Fe_3Al nanopowders were successfully synthesized. The as-prepared Fe-Al nanopowders exhibit a mean particle size around 100 nm. The mean Al concentration of the Fe-Al alloy is 31.0 ± 2.4 at.% which is acceptable to obtain the $D0_3$-ordered phase. Additionally, the $D0_3$-ordering can be significantly enhanced through the implementation of a suitable post-annealing treatment. The results prove that thermal plasma as a high-throughput process for the synthesis of chemically ordered magnetic nanopowders.

Keywords: thermal plasma, magnetic nanopowder, Fe_3Al, $D0_3$-ordering, energy-harvesting.

1. Introduction

Magnetic nanopowders (MNPs) have emerged as a topic of great interest in the scientific community due to their potential applications in various fields such as energy harvesting [1], 3D printing [2], catalysis [3], drug delivery [4], and biosensing [5]. Despite the various synthesis methods (such as wet chemical methods, mechanical grinding, and sputtering) used to produce MNPs, achieving the desired nanostructure remains a significant challenge [6]. A promising approach that has recently gained attention is thermal plasma synthesis to produce MNPs. In this study, we focused on synthesizing Fe_3Al-based nanopowders using a low oxygen induction thermal plasma (LO-ITP) system we recently developed [7]. Fe_3Al alloys have been well-studied due to their low material cost, good wear corrosion, sulfidation, and oxidation resistance compared to traditional stainless steels [8]. Recently, there has been a renewed interest in the $D0_3$-ordered Fe_3Al alloy due to its high Nernst thermopower coefficient, making it a suitable candidate for thermoelectric and energy harvesting applications [9]. Our promising results suggest that thermal plasma synthesis could be a high-throughput approach for the massive production of desired chemical-ordered nanopowders for micro-energy-harvesting applications and paves new opportunities for other fields such as additive manufacturing.

2. Experimental

Fe (3~5 μm, purity of 99.9%, Kojundo Chemical Lab. Co., Ltd., Japan) and Al (3~5 μm, purity of 99.9%, Kojundo Chemical Lab. Co., Ltd., Japan) powders were used as the starting materials and mixed with a mole ratio of Fe:Al = 70:30. The ITP was generated using a RF generator (TP-40020NPS, JEOL Ltd.) with a power of 6 kW and a frequency of 13.56 MHz. G1 grade Ar gas (purity 99.99995%) was used as the plasma gas with a flow rate of 35 L/min. The starting mixed powder was fed into the plasma in the gas stream at a flow rate of 3 L/min using a powder feeding system (TP-99010FDR, JEOL Ltd.). The as-prepared Fe-Al MNPs were then post-annealed at 500 °C under an Ar gas flow for 3 h to promote the $D0_3$-ordering. The detailed microstructure analysis was performed with X-ray diffraction (XRD), scanning electron microscope (SEM) and an atomic-resolution analytical electron microscope JEM-ARM200F. To prevent oxidation, most of the experiments and evaluations in this work were carried out under a low oxygen atmosphere (glovebox) without exposure to the atmosphere, except SEM and high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) characterizations.

3. Results and discussion

The mean particle size of the obtained Fe-Al alloy MNPs is around 100 nm. It should be noticed that by changing the quenching gas flow rate during the ITP process [10], one can tune the obtained particle size for different purposes. Elements mapping based energy dispersive X-ray spectroscopy (not shown here) demonstrates a homogenous Fe-Al composition distribution within and among the nanopowders which reveals a sufficient interdiffusion during the solidification process of the thermal plasma synthesis.

Fig. 1 shows the composition distribution of the as-prepared Fe-Al alloy MNPs which was analysis based on the count of 50 particles arbitrarily selected from the EDX elements mapping. The bin of the histogram was set to 2.0 at.%. The curve is the Gaussian fitting to the obtained histogram. According to the fitting, the mean value of Al concentration is 31.0 at.% with a standard deviation of 2.4 at.%. This result indicates that the Fe-Al alloy powder has a larger composition distribution than that of the Fe-Co alloy nanopowder [11], while a smaller composition distribution than that of the Ni-Cu alloy nanopowder [12] prepared also by the ITP process. According to the Fe-Al phase diagram, this composition deviation is acceptable to obtain the $D0_3$-ordered phase, although a perfect order phase is sacrificed.

According to the XRD results (not shown here), the as-prepared Fe-Al alloy MNPs contain both disordered A1(γ-phase) and ordered $D0_3$ phases. The degree of the $D0_3$-ordering can be significantly enhanced through post-annealing process under 500°C while maintaining the nanopowder feature without introducing serious coarsening. However, annealing at a temperature higher than 600°C will somehow trigger the coarsening of the nanopowders and degradation $D0_3$-ordering.

The $D0_3$-ordered structure of the post-annealed (500 °C × 3 hours) Fe-Al alloy MNPs was later confirmed via the detailed microstructure characterization. **Fig. 2** demonstrates the HAADF-STEM image of the post-annealed Fe-Al alloy MNPs. With the atomic-resolution HAADF-STEM image along [110] zone direction of one representative Fe_3Al MNPs, one can clearly detect a periodical ordered structure. The atoms with brighter contrast in the image are Fe atoms while the darker ones are Al atoms. Furthermore, the visible four-fold 111 superlattice reflections in the corresponding FFT image (inset of **Fig. 2**) indicate the formation of the $D0_3$-type ordered phase in the post-annealed Fe_3Al nanopowders. This is the first experimental demonstration of the synthesis of $D0_3$-ordered Fe_3Al nanopowders by the thermal plasma process.

4. Conclusions

In this work, we successfully synthesized the $D0_3$-ordered Fe_3Al nanopowders through a unique low oxygen induction thermal plasma system. Furthermore, this approach allows one to control the particle size of the final nanopowders by tuning the quenching rate during the thermal plasma process. Finally, appropriate post-annealing treatment can dramatically prompt the $D0_3$-ordering while avoiding the serious coarsening of the Fe_3Al magnetic nanopowders. All these merits make the proposed thermal plasma process a promising approach to the synthesis of chemically ordered nanopowders for its potential magnetothermal energy conversion and additive manufacturing applications.

5. References

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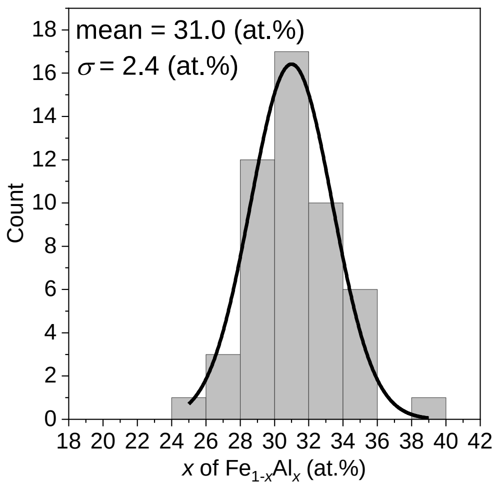

Fig. 1. Composition distribution histogram of the as-prepared Fe-Al alloy magnetic nanopowders.

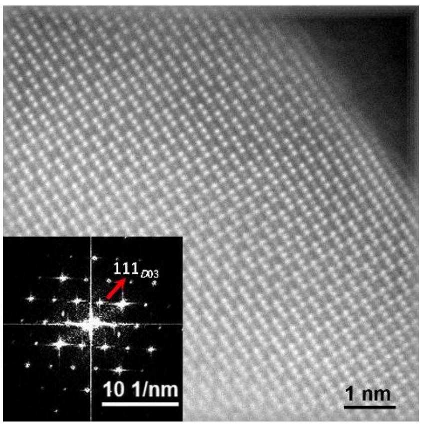

Fig. 2. Atomic resolution HAADF-STEM image of individual post-annealed Fe_3Al nanopowder with $D0_3$-ordering structure. Inset is the corresponding nanobeam diffraction pattern confirming the $D0_3$-ordering.