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One-step synthesis of γ-Fe_2O_3 nanoparticles by laser ablation

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Abstract

A Nd:YAG pulsed laser was used to ablate a 0.5-mm-diameter iron wire in a sealed chamber in a mixed gas flux of N_2 and O_2 to generate pure γ-Fe_2O_3 nanoparticles at atmospheric pressure. Structural characteristics and sizes of the prepared nanoparticles were determined by X-ray diffraction and TEM. The effects of laser power density, total mixed gas pressure and the oxygen ratio on the mean particle size were investigated, respectively. The results showed that the mean particle size decreased with the increase of the laser power density, total gas pressure and the oxygen ratio, respectively. Besides, the nanoparticle formation mechanism by laser ablation of iron wires was also discussed.

Keywords: Maghemite nanoparticles; Laser ablation; Nanoparticle size; Formation mechanism

1. Introduction

Maghemite (γ-Fe_2O_3) particles are the material most widely used for recording applications. Nanometer-sized maghemite particles show many distinct properties from their bulk materials or microparticles for high surface-to-volume ratio. The research on γ-Fe_2O_3 nanoparticle preparation and its applications have attracted great attention among researchers [1,2], especially on new synthesis methods of γ-Fe_2O_3. Up to now, the main technique of preparing γ-Fe_2O_3 nanoparticles is based on chemical reactions with different ferrous solutions. By this way, two or more steps are needed to get nanoparticles and extra impurities are introduced easily, which may decrease the preparation efficiency and result in the poorer magnetic property of the γ-Fe_2O_3 nanoparticles [3–5]. In recent years, some studies have shown that maghemite nanoparticles can be synthesized by other techniques, such as ball-milling method [6]. But this method also cannot keep clear of the impurity, similar to the chemical solution methods. On the other hand, physical gas-phase methods have shown good virtues for preparing small size and pure nanoparticles in the past years. Up to now, however, few reports about γ-Fe_2O_3 nanoparticles preparation by physical gas-phase methods were presented.

As a physical gas-phase method for preparing nanoparticles, pulsed laser ablation has become a popular method to prepare high-purity and ultra-fine nanoparticles of any composition. Nanoparticles of metals, intermetallic compounds, simple and complex oxides can be prepared easily by this method by controlling laser processing parameters and ambient gas pressure [7].

Now, we have realized one-step synthesis of high-purity γ-Fe_2O_3 nanoparticle by laser ablation of a tiny iron wire in a flowing mixed gas of N_2 and O_2 at atmospheric pressure. In this paper, we aim to introduce this new method and investigate the effects of laser power density, total mixed gas pressure and the oxygen ratio on the mean particle size. Besides, the nanoparticle formation mechanism was discussed.

2. Experimental details

The experimental configuration is depicted in Fig. 1. A metal wire is sent through the pipe furnace to the reaction area by an automatic wire-feeding system, and a pulsed Nd:YAG laser beam is focused by a plano-convex lens to the tip of the wire at normal incidence with a diameter of 0.5 mm. If necessary, the pipe resistance furnace with a power of 1 kW is used to preheat the wire to any designed temperature with a maximum temperature of 800 °C by an automatic temperature-control system. A special designed fixture is used to ensure that the wire can pass through smoothly and be irradiated accurately

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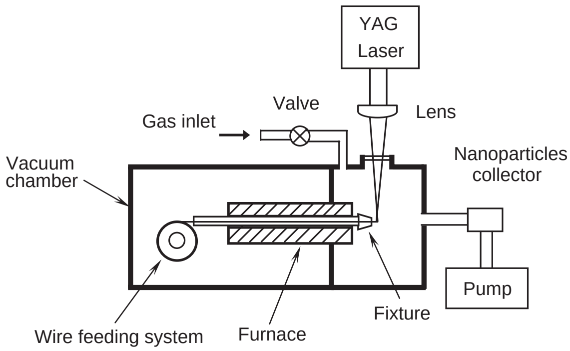

Fig. 1. Experimental configuration for nanoparticles preparation.

on the wire tip by the laser beam all the time. Furthermore, monotype or compound nanoparticles can be generated and collected with inert or reactive carrier gas with the same apparatus.

In our experiments, an iron wire with a diameter of 0.5 mm was used as the starting material for nanoparticle preparation. Pulsed laser parameters are as follows: laser wavelength 1064 nm, laser pulse width 0.3–20 ms, repetition rate 5–150 Hz, and average laser power 400 W. A mixture of N_2 (99.99%) and O_2 was used as a continuous reactive carrier gas during the period of nanoparticle preparation. As the wire ablated by laser pulses, the flowing mixed gas carried the fabricated nanoparticles into the collector by the help of a vacuum pump.

X-ray diffraction was used to determine the structure of the prepared nanoparticles. Specimens for TEM analysis were prepared by first dispersing some nanoparticles in ethanol, and then placing a drop on a carbon film carried on a 3-mm-diameter Cu grid followed by drying in air. A Philips CM12 transmission electron microscope (TEM) was used to observe the prepared nanoparticles and generate a sequence of micrographs of nanoparticles for each process condition. These micrographs were analyzed by an image processing software to determine the size and size distribution of the nanoparticles.

3. Results and discussion

In our experiments, all the produced powder were found to be γ-Fe_2O_3 by X-ray diffraction when the partial pressure of O_2 was above 0.01 MPa, which was also confirmed by analyses of the electron diffraction and X-ray photoelectron spectroscopy (the patterns are not given here). The X-ray diffraction pattern of the prepared γ-Fe_2O_3 nanoparticles is shown in Fig. 2. Fig. 3 shows a typical TEM micrograph of the γ-Fe_2O_3 nanoparticles, and it can be seen that the nanoparticles are basically of spherical shape and form chain-like structure due to their magnetism. Further TEM observation demonstrated that there are no big differences among the morphologies of the γ-Fe_2O_3 nanoparticles prepared at different processing conditions. In our statistics of the nanoparticles, we found that the particle sizes are basically homogeneous. The particle sizes ranged in diameters from 5 to 90 nm, and only a very small number of nanoparticles with the diameters of 50–90 nm were found. Especially, no nanoparticles larger than 90 nm were found in all TEM micrographs.

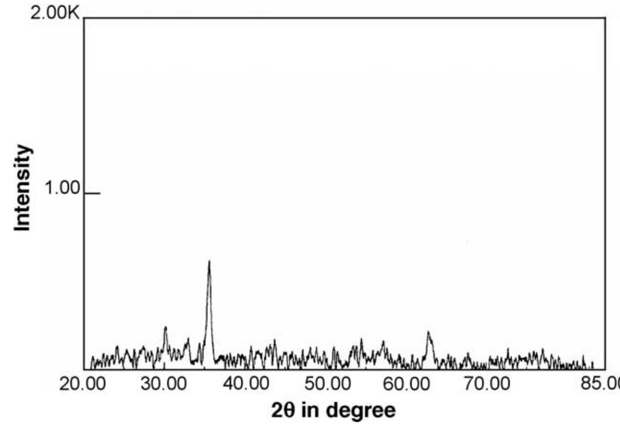

Fig. 2. XRD pattern for the prepared γ-Fe_2O_3 nanoparticles.

Fig. 4 shows the γ-Fe_2O_3 nanoparticle size statistics for a range of laser power densities at a constant mixed gas of 0.18 MPa N_2 and 0.02 MPa O_2. The mean particle diameter decreases gradually with increasing laser power density, which is consistent with the result on laser ablation of micropheres reported by Juang et al. [8], but is opposite to the results on laser ablation of bulk materials where the particle size increases with increasing laser power density [9,10]. Furthermore, the standard deviations at different conditions are nearly identical in value, which makes the change of the plot of S.D./Mean (standard deviation/mean diameter) be opposite to that of the mean diameter.

Except the laser processing parameters, the carrier gas pressure is another important parameter to control the mean diameter of nanoparticles. According to Figs. 5 and 6, the particle size of γ-Fe_2O_3 nanoparticles can easily be controlled by varying the total ambient pressure or the partial pressure of N_2 or O_2. In Fig. 5, the mean particle size of the prepared γ-Fe_2O_3 nanoparticles decreases with increasing total pressure of the mixed gas when the oxygen pressure is kept at 0.02 MPa. This result is similar to many previous reports [11,12], but is opposite to those where the preparation of nanoparticles took place in static buffer gases [7]. The difference is mainly due to the difference of gas movement mode. In our experiments, the

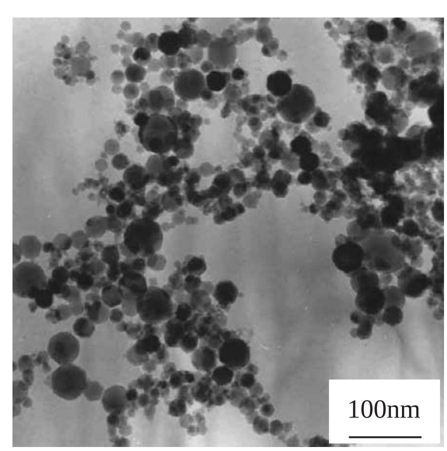

Fig. 3. TEM micrograph of γ-Fe_2O_3 nanoparticles at a laser power density of 6.8×10^6 W/cm^2 and a mixed gas of 0.02 MPa O_2 and 0.13 MPa N_2.