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Characterization of the magnetic Fe_3O_4 nanoparticles prepared by gamma irradiation

Tran Minh Quynh, Nguyen Van Binh, Nguyen Quang Long, Hoang Dang Sang

Hanoi Irradiation Center, Vietnam Atomic Energy Institute,
No.5, Minh Khai, Tu Liem, Hanoi

Abstract: In this study, gamma irradiation treatment has been applied for preparation of the magnetic nanoparticles from iron(III) chloride ($FeCl_3$). Structure of the resulting black powders were characterized with X-ray diffraction (XRD). The results revealed that the products prepared by gamma irradiation were mainly magnetite (Fe_3O_4) instead of maghemite (Fe_2O_3). The basic characteristics of the magnetite nanoparticles such as their particle size and magnetic properties were investigated using scanning electron microscope (SEM) and vibrating sample magnetometer (VSM). The nanoparticles showed superparamagnetism with the saturated magnetization of about 48.6 emu/g. Influences of radiation conditions on radiation preparation of magnetite nanoparticles and their properties were also discussed. Reaction yield increased, while average diameter of the obtained nanoparticles reduced from 46 to 17 nm depended on the absorbed dose.

Keywords: *gamma irradiation, nanoparticles, magnetization, X-ray diffraction*

I. INTRODUCTION

Magnetic Fe_3O_4 nanoparticles have been studied and applied in many various fields due to their superparamagnetic properties as well as their responsibility to surrounding fields. Several methods have been developed for preparation of magnetic nanoparticles such as reduction of hematite, the mineral form of iron(III) oxide (Fe_2O_3) by strong reductants [1], co-precipitation of admixture solutions containing ferrous and ferric salts, oxidation of the ferrous hydroxide gels using strong oxidants, thermal decomposition, sol-gel reactions…[2-4]. However, the obtained nanoparticles are rather large and inhomogeneous in size, limited their application in practice. During preparation of magnetic nanoparticle, the monodisperse size should be controlled due to the properties of the nanoparticles much depend on their dimension. Recently, gamma irradiation treatment has been developed for preparation the magnetic nanoparticles with more homogenous size [5]. This method can produce the products without any chemical residues, so that the obtained magnetic nanoparticles can be favourably used as biomaterials.

The magnetite nanoparticles with superparamagnetic behavior can be applied for many fields from biomedicine including of magnetic resonance imaging (MRI), immunoassay, hyperthermia, drug delivery carriers and cell separation to environment such as removal of pollutants from wastewater [4]. In this study, the gamma irradiation has

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been applied for preparation of the Fe_3O_4 nanoparticles from $FeCl_3$ solutions, and the effects of radiation conditions on the process were studied. The size, structure and magnetic properties of the obtained nanoparticles were characterized by SEM, XRD and vibrating sample magnetometer (VSM).

II. MATERIALS AND METHODS

2.1. Materials

$FeCl_3.6H_2O$, NH_3OH and other popular chemicals were bought from Xilong Chemical Co. Ltd., China. Isopropanol and other organic solvents were purchased from Dae Jung Chemicals and Metals. Co. Ltd. Korea.

2.2. Radiation preparation of magnetite (Fe_3O_4) nanoparticles.

Radiation preparation of magnetite nanoparticles includes two phases as reported by Wang et al [5]. Firstly, ferric hydroxide ($FeOOH.H_2O$) sols (yellow) were prepared by slowly dropping 50 mL NH_3OH 0.4 M into 50 mL $FeCl_3$ solution of 6 % (the molar ratio about 1.8 for NH_3 compared to Fe^{3+}) during stirring. Stirring continuously until the brown-red sols containing very small solid particles was formed, 20 mL isopropanol was added to the solution of β-FeOOH for OH scavenging. Secondly, the resulting sol was irradiated at various radiation doses ranging from 10-50 kGy under gamma Co-60 source at Hanoi Irradiation Center. After that, alkaline solution (NaOH 10%) was added to the irradiated solution for completely precipitation. The precipitates were separated, washed and dried under vacuum at 60°C for 6 hours. Finally, the products were ground into small size, and the obtained black magnetite nanoparticles were kept in a desiccator in order to avoid oxidation.

2.3. Measurements

The final products were characterized by using X-ray diffraction equipped with CuKα radiation (XRD, D8 Advance Bruker, Germany), scanning electron microscope (SEM, Hitachi, Japan) and vibrating sample magnetometer (VSM, DMS 880, USA). The reaction yield was determined by mass method. Average diameter and influence of radiation dose on the resulting nanoparticles was determined using their SEM images.

III. RESULTS AND DISCUSSIONS

3.1. Radiation preparation of magnetic nanoparticles

The results showed the black magnetite Fe_3O_4 nanoparticles can be easily formed during gamma irradiation of ferric hydroxide sol. However, the resulting precipitate formed by gamma irradiation at low radiation dose containing yellowish products of FeOOH as observed from Figure 1. Figure 2 shows XRD pattern of the magnetic nanoparticles prepared by gamma irradiation at dose of 40 kGy. The standard pattern of magnetite Fe_3O_4, which taken from library of X-ray Diffractometer, was also presented. It can be found that all diffraction peaks are corresponding to the standard Fe_3O_4. However, there are some noises in XRD pattern may due to the presence of other products existing in the precipitate. Moreover, the brown-red and yellowish dusts, the characteristic color of maghemite FeOOH can be also observed in Figure 1b, suggested that the precipitate is mainly magnetite with a few maghemite

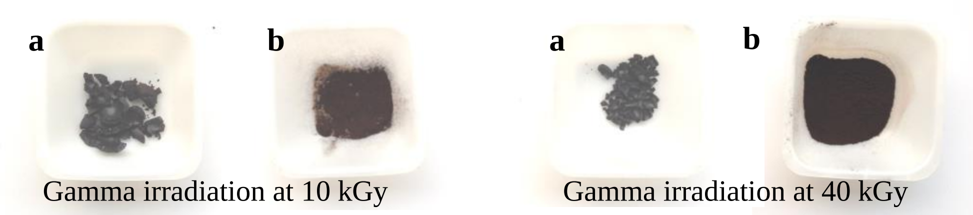

Fig.1. Photographs of the precipitate products of Fe_3O_4 (a), and Fe_3O_4 nanoparticles (b) prepared by irradiation method

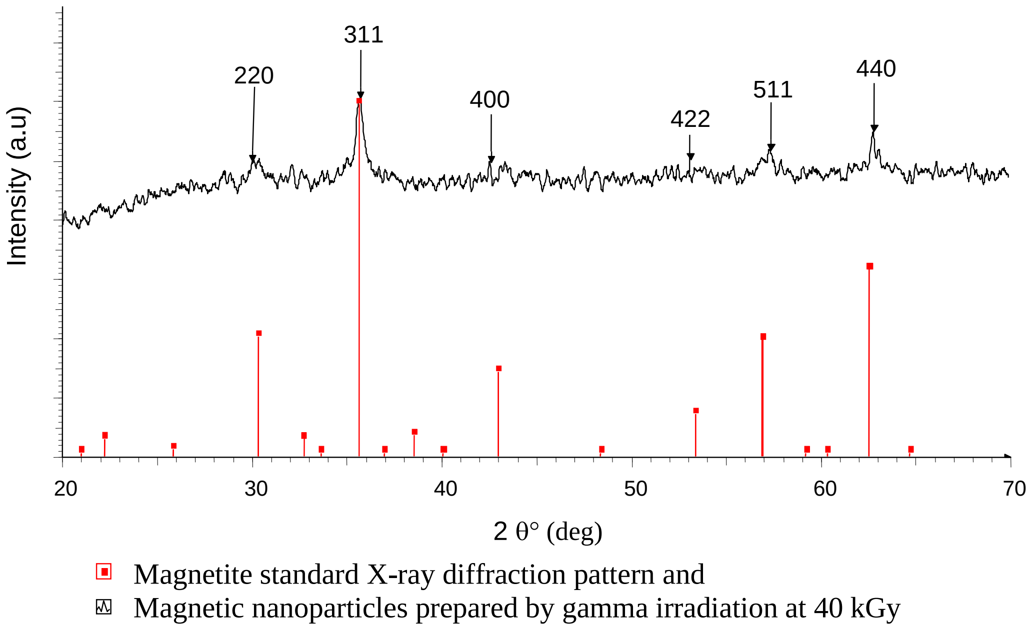

Fig.2. XRD patterns of the Fe_3O_4 nanoparticles prepared by irradiation method

3.2. Effect of radiation dose on the preparation and the size of Fe_3O_4 nanoparticles

In this study, the yield of radiation preparation of the magnetic iron oxide Fe_3O_4 is defined as the molar ratio of iron element in the product to that in the initial material of $FeCl_3$ used for the sols preparation. The average size of the resulting magnetite nanoparticles is determined and calculated with radiation dose from their SEM images. The results are show in Table I.

Table I. Preparation of magnetite nanoparticles by gamma radiation

Sample	Dose (kGy)	Yield* (%)	Average size (nm)
1	10	10.5	46
2	20	27.6	35
3	30	38.9	27
4	40	46.3	20
5	50	42.4	17

[*] The yield is defined as the molar ratio of the iron element in the product to that used in the initial hydroxide sols.

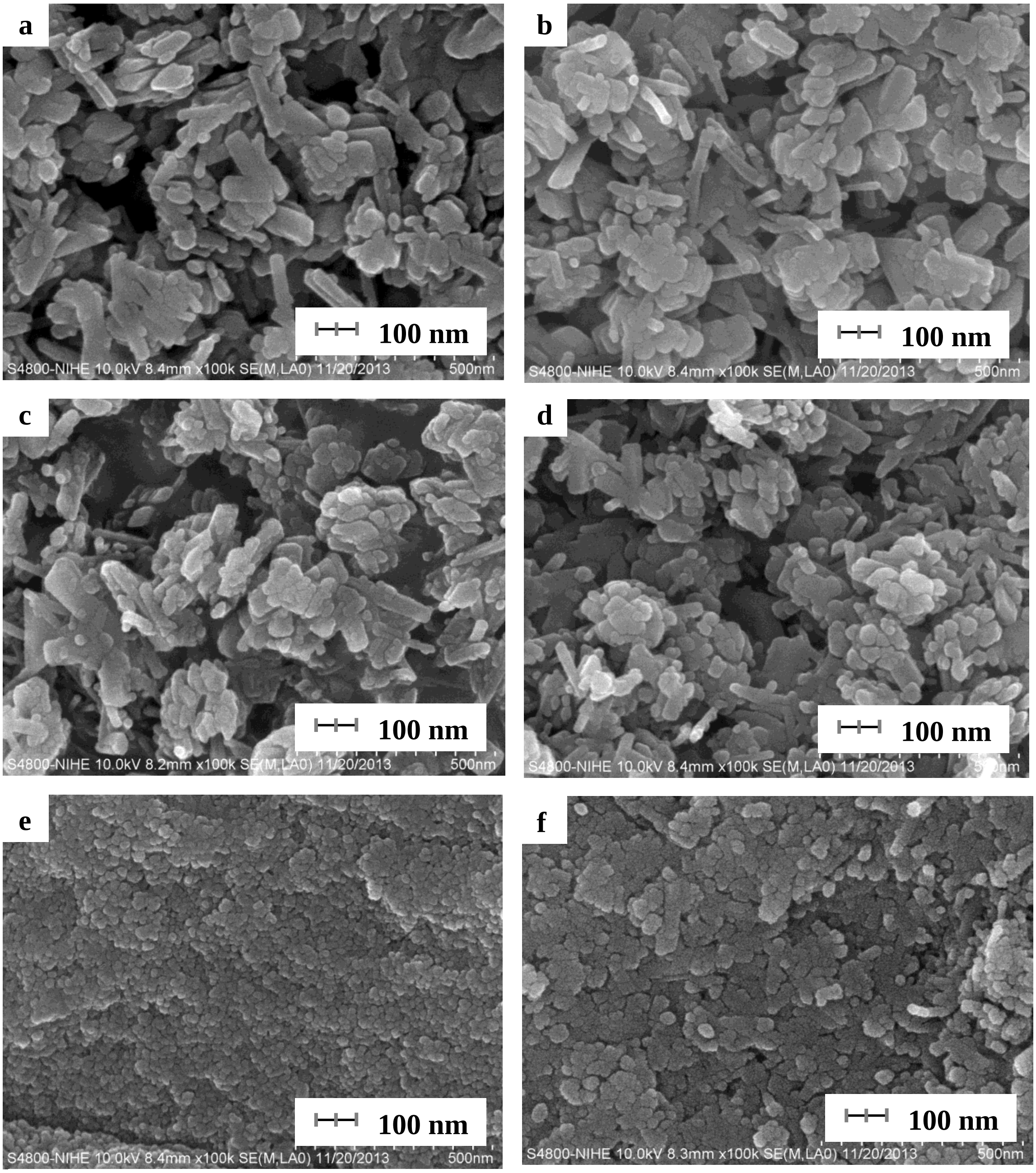

Fig.3. SEM photographs of the resulting precipitates from ferric hydroxide sols irradiated at 0 (a), 10 (b), 20 (c), 30 (d), 40 (e) and 50 kGy (f)

As presented in Table 1, the absorbed dose plays a key role for precipitation of magnetite nanoparticles. The conversion yield quickly increased with radiation dose to 40 kGy, then slightly reduced. During irradiation, the □ radiation provides the essential energy for the phase transition from □-FeOOH to □-FeOOH. And Fe_3O_4 was obtained by radiation-induced reduction of the □–FEOOH. However, the □-FeOOH is unstable compound that can be decomposed easily into a-Fe_2O_3 when heating, so that the hydroxide sols can also convert to Fe_2O_3 by irradiation at high dose as reported by Wang and Xin [3]. Moreover, the oxidation in air during storage may affect to the results [4]. Therefore, the dose of 40 kGy was selected as optimal dose for radiation preparation of magnetite nanoparticles. Though the yield of this radiation preparation was much smaller than that of the co-precipitation process, radiation method can improve the use of resulting magnetic nanoparticles beyond the applications in biomedicine.

Figure 3 shows the SEM images of the resulting nanoparticles from ferric hydroxide sols by gamma irradiation with different radiation doses. It can be found that spindle-shape structures representing for FeOOH particles resulting from ferric hydroxide, were changed into spherical nanoparticles of Fe_3O_4 during irradiation [5]. At low dose, the resulting precipitate containing both spindle structure of FeOOH and spherulite structure of Fe_3O_4. Precipitates consist of most fine magnetite nanoparticles were obtained by radiation processing of ferric hydroxide sols with dose higher than 40 kGy. Average diameter of the obtained magnetite nanoparticles reduced from 46 to 19 nm with irradiation dose as calculated from the photographs. Though smaller magnetic nanoparticles can be obtained by irradiation at 50 kGy, but they were aggregated. Therefore, the radiation dose of 40 kGy can be chosen as the most suitable dose for radiation preparation of magnetic nanoparticles from ferric chloride.

3.3. Magnetic properties of Fe_3O_4 nanoparticles

Figure 4 shows a typical magnetization curve of Fe_3O_4 nanoparticles obtained by gamma irradiation at 40 kGy. The hysteretic loop revealed superparamagnetic property of the nanoparticles. The saturated magnetization of the obtained Fe_3O_4 nanoparticles was about 48.6 $emu.g^{-1}$, lower than those prepared by co-precipitation methods [6]. There are no remanence and coercivity can be observed in the figure, suggested that the magnetic nanoparticles are superparamagnetic, i.e. responsiveness to an applied magnetic field

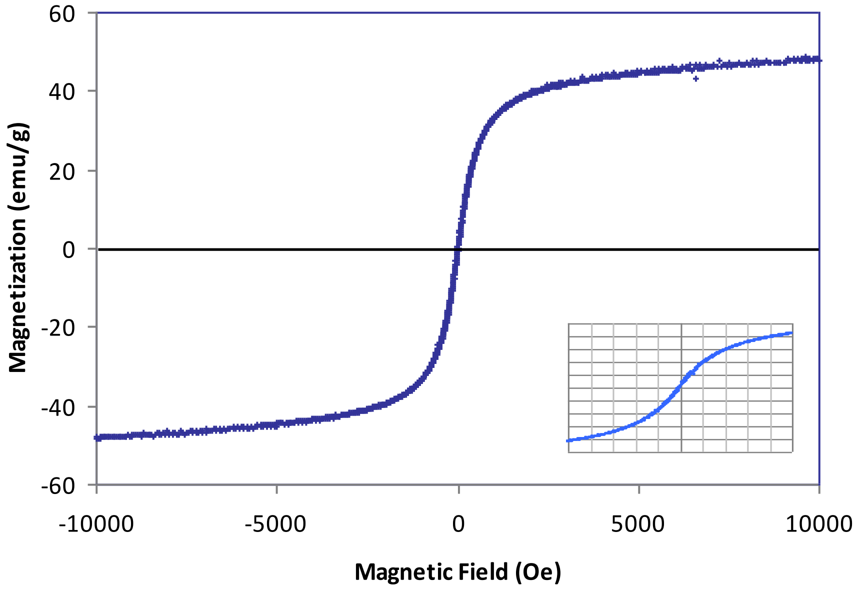

Fig 4. Magnetic hysteresis curve of Fe_3O_4 nanoparticles prepared by gamma irradiation at 40 kGy measured at room temperature

without retaining any magnetism after removal of the external field [7-9]. Therefore, the magnetic nanoparticles prepared by gamma irradiation can be used as targeting drug carriers.

IV. CONCLUSION

Homogenous magnetite Fe_3O_4 nanoparticles were obtained from iron (III) chloride solution by gamma irradiation method. The results indicated that the ferric hydroxide sols, then spindle shaped nanostrutures of FeOOH can be easily formed from the ferric solution. These nanostructures changed into the spherical particles of Fe_3O_4 by gamma irradiation. The conversion yield of magnetite nanoparticles increased, whereas their average size reduced with radiation dose, and the dose of 40 kGy can be chosen as the most suitable dose for the radiation preparation. XRD pattern revealed the resulting products mainly contain magnetite Fe_3O_4 with few impurities. The magnetic nanoparticles are superparamagnetic with saturated magnetization of 48.6 $emu.g^{-1}$, can be applied as magnetic responsive materials for further applications.

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