

Enhancement of the recoilless fraction in $R_2Fe_{10}Co_4Si_2$ ($R = Gd, Er, Y$) offstoichiometric compounds after hydrogenation

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Received 19 October 2007; received in revised form 15 January 2008; accepted 7 February 2008

Abstract

Samples of $R_2Fe_{10}Co_4Si_2$ ($R = Gd, Er$ and Y) solid solutions were prepared by arc melting and exposed to hydrogenation. Characterization of hydrogenation effects was done using Mössbauer spectroscopy. The values of the magnetic hyperfine fields corresponding to the four inequivalent lattice sites were found to increase after hydrogenation in all offstoichiometric compounds investigated. Slight changes in the relative site occupancies were observed. Transmission Mössbauer spectra were also recorded for absorbers consisting of the intermetallic and the stainless steel etalon. The recoilless fraction was derived from these spectra using the method developed by us. Our study is the first to evidence that the recoilless fraction is enhanced after hydrogenation.

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PACS: 76.80+y

Keywords: Magnetic materials; Intermetallics; Recoilless fraction; Mössbauer spectroscopy

1. Introduction

Hydrogenation of solid solutions of the type R_2Fe_{17} was found to increase the Curie temperature and develop the suitability of these materials for permanent magnet applications [1–4]. The use of substitutions in this structure led to a further enhancement of the ordering temperature in these compounds [5,6]. In the present paper we propose to study atomic architectures of the type $R_2Fe_{10}Co_4Si_2$ ($R = Gd, Er$ and Y) by transmission Mössbauer spectroscopy. We focus our attention on the effects of hydrogenation on the hyperfine magnetic fields and site occupancy. However, this study would not be complete without a discussion of hydrogenation effects on the recoilless fraction. It is possible to evidence these effects only due to our dual absorber method that we developed for the determination of the recoilless fraction [7–12].

2. Experimental

The samples of $R_2Fe_{10}Co_4Si_2$ ($R = Gd, Er$ and Y) were prepared by induction melting amounts of constituent elements in a water-cooled copper boat under a continuous flow of purified argon gas. The ingots were turned over and remelted several times to insure homogeneity. Then, the compounds were annealed at 1000 °C for 3 h in the boat.

To obtain structural and magnetic data on the hydrides, the compounds were exposed to high purity hydrogen at a pressure of about 60 atm, using conventional volumetric equipment. The hydrogen uptake was complete in a few minutes. The amount of hydrogen absorbed was calculated by noting the change of pressure in a closed system apparatus of known volume. The hydrides were then quenched in liquid nitrogen and the remnant gaseous hydrogen rapidly pumped away.

Mössbauer spectroscopy measurements were made at room temperature using a constant acceleration spectrometer. The 10 mCi gamma ray source was ^{57}Co in a Rh matrix, maintained at room temperature. All spectra were analyzed with the NORMOS program. For the

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determination of the recoilless fraction using our dual absorber method, the sample consists of the intermetallic sandwiched together with the stainless steel etalon and the recoilless fraction is determined using the relative areas of the component subspectra.

3. Results and discussion

Fig. 1(a) presents the room temperature transmission Mössbauer spectrum of $\text{Gd}_2\text{Fe}_{10}\text{Co}_4\text{Si}_2$ before hydrogenation. The hyperfine parameters extracted from this spectrum by least-squares fitting are summarized in Table 1. According to literature [5], $\text{Gd}_2\text{Fe}_{10}\text{Co}_4\text{Si}_2$ is rhombohedral (R_2Zn_{17} -type) and the inequivalent lattice sites are 6c, 9d, 18f and 18h. The spectrum was fitted with four sextets, corresponding to the four inequivalent lattice sites [3]. The sites are given in the order of decreasing the hyperfine magnetic field. We further refined the fitting procedure by

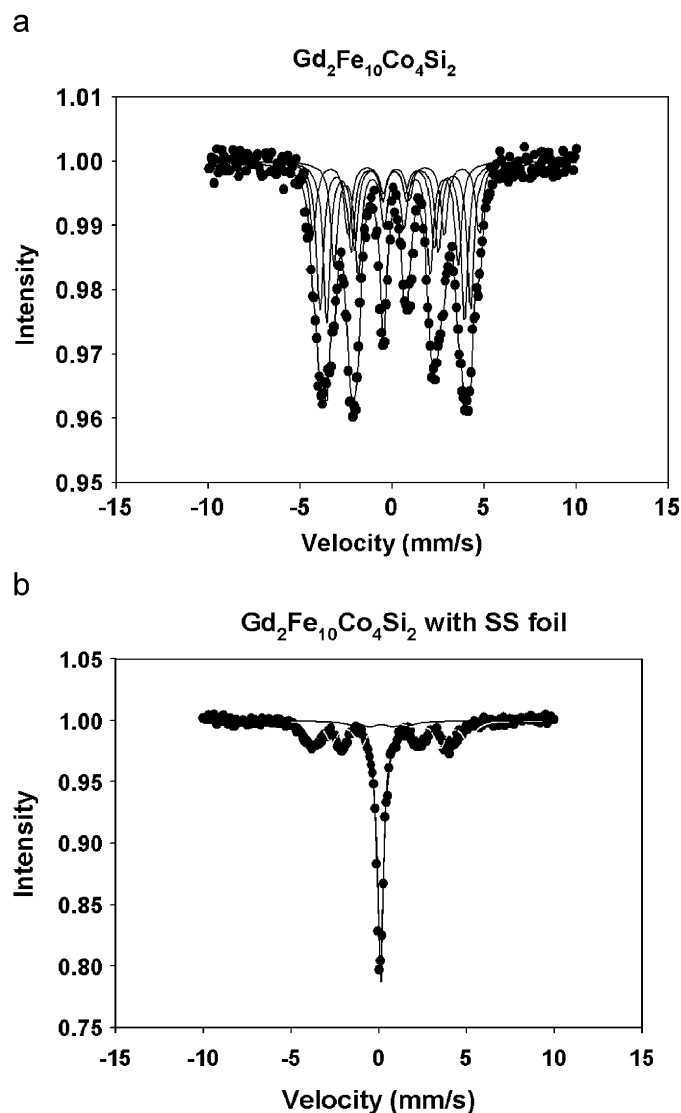


Fig. 1. Room temperature transmission Mössbauer spectra of $\text{Gd}_2\text{Fe}_{10}\text{Co}_4\text{Si}_2$ (a) as-obtained and (b) with the stainless steel foil.

Table 1
Hyperfine parameters of $\text{Gd}_2\text{Fe}_{10}\text{Co}_4\text{Si}_2$ before and after hydrogenation, with and without the stainless steel foil

| Sample | BHF (T) | QS (mm/s) | IS/Fe (mm/s) | A (%) |
|--------|------------|-------------|--------------|-----------|
| Gd | 28.03 | 0.153 | 0.201 | 18.2 |
| | 25.42 | 0.031 | 0.134 | 27.2 |
| | 23.27 | 0.070 | 0.129 | 27.3 |
| | 20.90 | 0.115 | 0.135 | 27.3 |
| Gd/SS | 24.17 | -0.030 | 0.014 | 40.4 |
| | | | -0.016 | 59.6 |
| GdH | 28.90 | -0.116 | 0.243 | 18.1 |
| | 25.56 | -0.162 | 0.149 | 27.3 |
| | 23.43 | -0.028 | 0.194 | 27.3 |
| | 20.92 | 0.002 | 0.242 | 27.3 |
| GdH/SS | 23.73 | -0.057 | 0.003 | 53.3 |
| | | | -0.019 | 46.7 |
| Errors | ± 0.05 | ± 0.002 | ± 0.002 | ± 0.5 |

BHF is the magnetic hyperfine field, QS is the quadrupole shift, IS is the isomer shift relative to iron and A is the relative area of subspectra.

Table 2
Hyperfine parameters of $\text{Er}_2\text{Fe}_{10}\text{Co}_4\text{Si}_2$ before and after hydrogenation, with and without the stainless steel foil

| Sample | BHF (T) | QS (mm/s) | IS/Fe (mm/s) | A (%) |
|--------|------------|-------------|--------------|-----------|
| Er | 28.11 | -0.088 | 0.173 | 18.0 |
| | 24.99 | -0.079 | 0.109 | 26.0 |
| | 22.76 | -0.039 | 0.134 | 28.0 |
| | 20.25 | 0.101 | 0.124 | 28.03 |
| Er/SS | 23.74 | -0.037 | -0.022 | 46.8 |
| | | | -0.023 | 53.23 |
| ErH | 28.15 | -0.095 | 0.217 | 18.8 |
| | 25.84 | -0.102 | 0.124 | 27.0 |
| | 23.40 | -0.127 | 0.161 | 27.1 |
| | 20.98 | 0.039 | 0.147 | 27.13 |
| ErH/SS | 23.73 | -0.078 | -0.003 | 49.0 |
| | | | -0.021 | 51.0 |
| Errors | ± 0.05 | ± 0.002 | ± 0.002 | ± 0.5 |

BHF is the magnetic hyperfine field, QS is the quadrupole shift, IS is the isomer shift relative to iron and A is the relative area of subspectra.

freeing the great majority of variables, initially constrained. Only the line widths are now fixed and the spectral areas were correlated to be in agreement with the crystallographic data (for instance, $A_1 < 20\%$, $A_2 < 30\%$, etc). This is the ideal fit and the new fitted variables are given in Tables 1–3. Fig. 1(b) shows the room temperature transmission Mössbauer spectrum of the $\text{Gd}_2\text{Fe}_{10}\text{Co}_4\text{Si}_2$ intermetallic recorded simultaneously with the spectrum of the stainless steel etalon. The deconvolution was performed using a sextet for the intermetallic and a singlet for the stainless steel foil. The average sextet for the intermetallic was used instead of the four-site model, because the four sextets are not well resolved in the compound spectra.

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