Structure and magnetic properties of layered \((\text{Fe}_x\text{Co}_{1-x})\text{Ta}_2\text{O}_6\) compounds

V.D. Mello, L.I. Zawislak, J.B. Marimon da Cunha, E.J. Kinast, J.B. Soares, C.A. dos Santos*

Instituto de Física–UFRGS, Av. Bento Gonçalves 9500, C.P. 15051-Campus do Vale, 91501-970 Porto Alegre, RS, Brazil

Abstract

Samples of \((\text{Fe}_x\text{Co}_{1-x})\text{Ta}_2\text{O}_6\) were prepared from pure \(\text{FeTa}_2\text{O}_6\) and \(\text{CoTa}_2\text{O}_6\). From X-ray diffraction (XRD) measurements followed by Rietveld refinement, it is demonstrated that the solid solution obeys the Vegard’s law. Mössbauer spectroscopy (MS) measurements at 4.2 and 300 K show that for \(x \geq 0.8\) the Fe\(\rightarrow\)Co substitution has no apparent effect on the hyperfine parameters. © 1999 Elsevier Science B.V. All rights reserved.

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In the tetragonal isomorphous \(\text{ATa}_2\text{O}_6\) compounds \((A = \text{Fe}, \text{Co}, \text{Ni})\), the sublattice formed by the 3d transition metal ions have the same symmetry as the Ni sublattice in \(\text{K}_2\text{NiF}_4\), the well-known two-dimensional Heisenberg antiferromagnet [1,2]. Recent Mössbauer spectroscopy (MS) measurements [3] have shown that from room temperature (RT) to about 15 K, \(\text{FeTa}_2\text{O}_6\) is clearly in the paramagnetic regime. The onset of long-range order is manifested near 10 K by the emerging hyperfine magnetic field. Magnetization curves [4], for temperatures between 1.4 and 5 K, display a noticeable change in slope, suggesting a spin-flop transition induced by a field higher than 10 T.

Similar results were obtained for \(\text{CoTa}_2\text{O}_6\) [2], but the magnetic structure, as suggested by powder neutron diffraction taken at 4.2 K [5], is much more complex than for \(\text{FeTa}_2\text{O}_6\). A tow-cone axis helical spin structure was proposed. Therefore, random mixtures of the type \((\text{Fe}_x\text{Co}_{1-x})\text{Ta}_2\text{O}_6\) are very appropriate to investigations on the effect of the competing anisotropies on the magnetic-phase diagram.

In the present work we report, for the first time, powder X-ray diffraction (XRD) and MS measurements on samples \((\text{Fe}_x\text{Co}_{1-x})\text{Ta}_2\text{O}_6\), with \(x = 0, 0.4, 0.8\) and 1. \(\text{FeTa}_2\text{O}_6\) was prepared in vacuum, as described in Ref. [4]. \(\text{CoTa}_2\text{O}_6\) was prepared in a similar way, but in air, instead of vacuum. For \((\text{Fe}_x\text{Co}_{1-x})\text{Ta}_2\text{O}_6\) samples, appropriate amounts of \(\text{FeTa}_2\text{O}_6\) and \(\text{CoTa}_2\text{O}_6\) were mixed and processed as for the \(\text{FeTa}_2\text{O}_6\) preparation. XRD patterns were obtained in a Bragg–Brentano parafocusing geometry, under experimental setup as described in Ref. [4]. Structural parameters were refined with the program Fullprof [6]. The MS spectra were obtained at room temperature (RT) and at 4.2 K using a conventional spectrometer. The hyperfine parameters were obtained by a least-squares procedure. \(^{57}\text{Co}\) in rhodium was used at room temperature as a source, with nominal activity of 50 mCi.

The XRD patterns for all the samples were indexed to the space group \(\text{P}4_2/\text{mmm}\), as expected for a trirutile structure. Typical pattern is shown in Fig. 1 for \(\text{Fe}_{0.4}\text{Co}_{0.6}\text{Ta}_2\text{O}_6\). The small Bragg factor obtained for all the Rietveld refinements and the absence of spurious reflections on the XRD diagrams are strong indications that we have prepared well-crystallised single-phase samples, with lattice parameters typical of the tapiolite–mososite system \((a \approx 0.47 \text{ nm}, \ c \approx 0.92 \text{ nm})\).
Fig. 1. Room temperature X-ray powder diffraction pattern for (Fe<sub>0.4</sub>Co<sub>0.6</sub>)Ta<sub>2</sub>O<sub>6</sub>.

Fig. 2. Variation in lattice parameter c with chemical composition.

c parameter is plotted as a function of chemical composition. The linear dependence reflects the Vegard’s law validity for solid solubility.

Another evidence of the good quality of the (Fe<sub>x</sub>Co<sub>1-x</sub>)Ta<sub>2</sub>O<sub>6</sub> samples comes from the MS measurements, as illustrated in Fig. 3 for the Fe<sub>0.8</sub>Co<sub>0.2</sub>Ta<sub>2</sub>O<sub>6</sub> sample at RT. All the spectra at RT were fitted to a quadrupole doublet with linewidth at half-height (\( \Gamma \)) in the range 0.26–0.27 mm/s. As the iron foil calibration resulted in \( \Gamma = 0.26 \) mm/s, it is reasonable to suppose no significant impurity effect on the Mössbauer spectra of the present samples. From the crystallographic data it becomes evident that Fe\(^{2+}\) (Co\(^{2+}\)) and Ta\(^{5+}\) cations are surrounded by O\(^{2-}\) octahedra, and that Fe–O(Ta–O) planes are well defined in the trirutile structure of (Fe<sub>x</sub>Co<sub>1-x</sub>)Ta<sub>2</sub>O<sub>6</sub>. Successive Fe–O(Co–O) planes are separated by two Ta–O planes. The hyperfine parameters are constant as a function of chemical composition for \( x \geq 0.8 \). This trend is observed at 300 and at 4.2 K. In conclusion, the tetragonal (Fe<sub>x</sub>Co<sub>1-x</sub>)Ta<sub>2</sub>O<sub>6</sub> system is a continuous solid solution, for which the effect of the Fe–Co substitution on the lattice parameters \( a \) and \( c \) is consistent with the Vegard’s law. On the other hand Fe–Co substitution have no effect on the hyperfine parameters for iron-rich samples.

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References