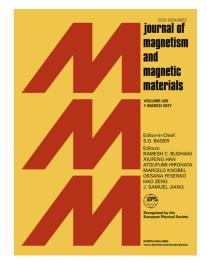
### Accepted Manuscript

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PII:	S0304-8853(17)31482-8
DOI:	https://doi.org/10.1016/j.jmmm.2017.10.105
Reference:	MAGMA 63321
To appear in:	Journal of Magnetism and Magnetic Materials
Received Date:	12 May 2017
Revised Date:	17 October 2017
Accepted Date:	26 October 2017



Please cite this article as: P. Mohanty, C.J. Sheppard, A.R.E. Prinsloo, W.D. Roos, L. Olivi, G. Aquilanti, Effect of cobalt substitution on the magnetic properties of nickel chromite, *Journal of Magnetism and Magnetic Materials* (2017), doi: https://doi.org/10.1016/j.jmmm.2017.10.105

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## ACCEPTED MANUSCRIPT

## Effect of cobalt substitution on the magnetic properties of nickel chromite

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#### ABSTRACT

It is of interest to study the magneto-structural coupling in geometrically frustrated antiferromagnets, where structural distortion elevates the ground state degeneracy, leading to a long-range magnetic order. In this regard a cubic spinel compound of the form  $AB_2O_4$  is currently drawing much attention, where A refers to tetrahedral and B to octahedral sites. In the particular case of NiCr<sub>2</sub>O<sub>4</sub> the material undergoes several structural phase transitions associated with the magnetic ordering. It is therefore necessary to study the magnetic behaviour of NiCr<sub>2</sub>O<sub>4</sub> by substituting either A or B sites, or both systematically with suitable cations. The current work therefore aims at the modification of magnetic properties of  $NiCr_2O_4$  by doping with  $Co^{2+}$  at A sites. In order to achieve the afore mentioned,  $(Ni_{1-x}Co_x)Cr_2O_4$  ( $0 \le x \le 1$ ) were prepared using chemical co-precipitation techniques. X-ray diffraction (XRD) results indicate that the samples are in the expected phase without any trace of Cr<sub>2</sub>O<sub>3</sub> impurities after calcination. Transmission electron microcopy (TEM) analyses of these samples show that the particles are mostly bipyramidal in shape, with sizes ranging from 50 nm to 100 nm. In the present study the ferrimagnetic transition temperatures  $(T_{\rm C})$  of the various samples were determined utilizing magnetization as function of temperature measurements.  $T_{\rm C}$  for NiCr<sub>2</sub>O<sub>4</sub> and CoCr<sub>2</sub>O<sub>4</sub> was determined to be  $82.4 \pm 0.8$  K and  $99.5 \pm 0.5$  K, respectively. These values are higher than those previously reported in the literature for both these compounds. Substitution of Ni by Co, results in an increase in the  $T_{\rm C}$ , giving values of 89.2  $\pm$  0.7 K and 90.6  $\pm$  0.9 K for (Ni<sub>0.5</sub>Co<sub>0.5</sub>)Cr<sub>2</sub>O<sub>4</sub> and (Ni<sub>0.25</sub>Co<sub>0.75</sub>)Cr<sub>2</sub>O<sub>4</sub>, respectively. The (Ni<sub>0.5</sub>Co<sub>0.5</sub>)Cr<sub>2</sub>O<sub>4</sub> sample demonstrated a high coercivity of  $3.6 \pm 0.1$  T and a shift in the hysteresis loop observed under field cooled measurement, not previously reported in literature. X-ray photoelectron spectroscopy (XPS) of (Ni<sub>0.5</sub>Co<sub>0.5</sub>)Cr<sub>2</sub>O<sub>4</sub> suggests that the oxidation states of Ni and Co are 2+, while that of Cr is 3+. In order to investigate the local structure around the cations, low temperature extended x-ray absorption fine structure (EXAFS) measurements were performed. From EXAFS it is confirmed that no redistribution of A and B site cations occur at low temperatures. Present findings demonstrate that the magnetic properties of NiCr<sub>2</sub>O<sub>4</sub> can easily and dramatically be modified by doping  $Co^{2+}$ at A sites.

Keywords: Chromites, Magnetic frustration, EXAFS.

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