Available online at www.sciencedirect.com

SCIENCE DIRECT®

Materials Research Bulletin 40 (2005) 840–849

Materials Research Bulletin

www.elsevier.com/locate/matresbu

The magnetoelectric perovskite Sr_2CoMoO_6: An insight from neutron powder diffraction

S.A. Ivanov [a,*], S.-G. Eriksson [b], R. Tellgren [c], H. Rundlöf [c], M. Tseggai [c]

[a] Department of Inorganic Materials, X-ray laboratory, Karpov' Institute of Physical Chemistry, Ul.Vorontsovo pole 10, 105064 Moscow K-64, Russia

[b] Department of Inorganic Chemistry, University of Gothenburg, Sweden

[c] Department of Materials Chemistry, The Ångstrom Laboratory, University of Uppsala, Uppsala, Sweden

Received 26 November 2004; accepted 18 January 2005

Abstract

A study of the crystallographic and magnetic structure of the double perovskite Sr_2CoMoO_6 (SCMO) has been carried out on a polycrystalline sample using neutron powder diffraction (NPD) data between 10 and 700 K. An analysis of the NPD patterns at room temperature has shown that this compound crystallises in the tetragonal space group $I4/m$ with a = 5.5616(1) Å and c = 7.9470(2) Å and has a 1:1 ordered arrangement of Co and Mo at the B-sites. This compound undergoes a $I4/m \rightarrow Fm3m$ improper ferroelectric phase transition near 560 K. A low-temperature antiferromagnetic ordering (below T_N = 36 K) has been followed from sequential NPD data analysis. The antiferromagnetic structure is defined by the propagation vector k = (1/2, 0, 1/2). In addition to the obtained experimental results on magnetic and electric properties some aspects of magnetoelectricity in this perovskite are also discussed.

Keywords: A. Ceramics; C. Neutron scattering; D. Crystal structure; D. Magnetic structure

* Corresponding author. Tel.: +7 95 917 31 46; fax: +7 95 975 24 50.

E-mail addresses: ivan@cc.nifhi.ac.ru, sergey.ivanov@studsvik.uu.se (S.A. Ivanov), stene@inoc.chalmers.se, sten.eriksson@studsvik.uu.se (S.-G. Eriksson), rte@kemi.uu.se (R. Tellgren), rundlof@studsvik.uu.se (H. Rundlöf).

0025-5408/$ – see front matter

doi:10.1016/j.materresbull.2005.01.012

1. Introduction

Magnetoelectric materials, simultaneously manifesting ferroelectric and ferromagnetic properties, have been interesting subjects of study not only for their possible application in electronic devices but also from the viewpoint of fundamental materials science [1]. At present time, the number of candidate materials is strongly limited and the effects are typically too small to be useful in applications [2]. It is very important to understand under which thermodynamic conditions and chemical environment the coexistence of magnetic and ferroelectric orderings can take place. It was shown [2] that ferroelectricity and magnetism are rarely found in the same compound. Much experimental work still needs to be done in order to reach a clear understanding of this coexistence and eventually to create new such compositions. Preliminary studies of double perovskites seem to indicate that the occurrence of magnetoelectric properties is a common feature in some of them [1].

Despite the recent progress in understanding the physics of the double perovskites the origin of magnetism and ferroic polar distortion is still controversial. But it is clear now that these physical properties are very sensitive to even small structural changes. Sr_2CoMoO_6 (SCMO) has attracted our attention with the aim of finding a new magnetoelectric perovskite. This complex metal oxide was synthesised for the first time almost 45 years ago and then forgotten for many years. It belongs to the class of perovskite compounds, which simultaneously can exhibit the desired long-range ordering of electrical and magnetic dipoles [3–5]. This double perovskite was first prepared and studied by Galasso [3]. Later it was reported to be an antiferromagnetic with T_N = 34–37 K [4–7] and an improper ferroelectric with T_C = 563 K [8]. The crystal structure at room temperature has been reported several times [3–8], but there are some differences in the structural details reported by different authors and still many points remain unclear. X-ray diffraction was the main technique used in these works to elucidate the structure of SCMO. Because the neutron powder diffraction (NPD) technique is more sensitive than X-ray techniques for structural studies of magnetoelectric oxides with strong cubic pseudosymmetry, we have undertaken NPD studies on this compound. The main purpose of this research is to resolve the discrepancy in the literature on SCMO and to determine the temperature evolution of the nuclear and magnetic structures.

2. Experimental

According to the procedure described in [8], a high quality ceramic Sr_2CoMoO_6 sample was prepared by mixing appropriate amounts of oxides or carbonates. The mixture was ground in ethanol and then sintered in air at 1523 K for 24 h, pressed into pellet and fired at 1623 K for 10 h with one intermediate grinding. The sample was characterised by X-ray powder diffraction (XRD diffractometer URD-63, Cu Kα radiation) for phase identification and to check phase purity. The prepared sample was a single phase; no impurity lines could be detected.

The chemical composition of the prepared ceramic SCMO sample was analysed by energy-dispersive spectroscopy (EDS) coupled to a transmission electron microscope. The analyses performed on several particles showed that the studied sample has close to the expected cation ratio, that was estimated as $Sr_{2.01(2)}Co_{0.98(2)}Mo_{1.01(2)}O_6$.

Magnetic susceptibility was measured with a Quantum Design SQUID magnetometer at an applied field of 10 Oe in the temperature range 5–400 K. Zero field cooled (ZFC) measurements were performed by cooling the samples to 10 K, applying the field, and measuring the magnetisation as the sample was