

## ABSTRACT

Double perovskite oxides are becoming more and more important in various technological applications. There is a need to improve our detailed understanding of the structural and magnetic properties of such materials and how the atomic structure is related to macroscopic physical properties. Neutron diffraction in combination with Rietveld refinements and magnetization measurements has proven to be a very useful tool in the study of structural and magnetic properties of double perovskites. The structure and physical properties of this type of double perovskites depend considerably on the size, valence and order of the  $A$  and  $B$  site cations. In this thesis,  $A_2\text{MnBO}_6$  and  $A_2\text{FeBO}_6$  ( $A = \text{Ca, Sr, Ba, La}$ ;  $B = \text{W, Mo, Cr}$ ) systems were chosen for investigation to gain an understanding of the structural and magnetic details, and the concentration dependence of  $A$ -site cations.

Polycrystalline samples were prepared by solid-state sintering of metal oxides and carbonates. The samples were preliminarily characterised by X-ray powder diffraction in order to examine the purity of the products solid solubility, and to obtain preliminary information about space group symmetry. Then neutron powder diffraction and magnetization measurements were carried out to study the structural and magnetic properties. Other characterization methods such as ED, HRTEM, SEM, EDX were performed when necessary.

The room and low temperature structures of these compounds were refined by the Rietveld method to derive the structural details and correlation between the compounds. The structure changes from monoclinic through tetragonal to cubic with the change of the  $A$ -site cation as well as with the change of temperature. A structural and magnetic phase diagram is proposed for  $A_2\text{MnWO}_6$  (Ca, Sr, Ba) compounds from the study. All samples were  $B$ -site ordered except  $\text{La}_{2-x}\text{Sr}_x\text{FeCrO}_6$  ( $x = 0.0, 0.33, 0.67, 1.0$ ). The magnetic transition temperature depends on the composition, bond distances, interaction and ordering. A decrease of the paramagnetic to antiferromagnetic transition temperature with an increase in the  $A$ -site cation radius was also observed in the compositions  $A_2\text{MnWO}_6$  (Ca, Sr, Ba) and  $A_2\text{FeWO}_6$  (Sr, Ba) from the present investigation. Bond valence sum calculations indicate that the charge distribution between Mn, and W in  $A_2\text{MnWO}_6$  (Ca, Sr, Ba) were  $\text{Mn}^{2+}$  and  $\text{W}^{6+}$ , and in  $A_2\text{FeWO}_6$  (Sr, Ba) and  $A_2\text{MnMoO}_6$  (Sr, Ba), there were mixed valence states between  $\text{Fe}^{2+}/\text{Fe}^{3+}$ ,  $\text{W}^{6+}/\text{W}^{5+}$   $\text{Mn}^{2+}/\text{Mn}^{3+}$ , and  $\text{Mo}^{6+}/\text{Mo}^{5+}$ , respectively.

*Key words:* Double perovskite, neutron diffraction, crystal structure, magnetic structure, magnetization measurement