

## ABSTRACT

The present research work reports experimental studies on the phase formation, magnetostructural and magnetocaloric (MC) properties of a novel series of ferromagnetic shape memory materials of off-stoichiometric Heusler alloys  $\text{Ni}_2\text{MnSn}$  in form of a cylindrical disc. The aim is two fold; one from the applied point of view and the other from fundamental aspects. In the application part, the main objective is to develop a magnetic refrigerant with large MC properties near room temperature, whereas in the other part, the motive is to develop basic understanding of underlying processes of the structural, thermomagnetic, magneto-transport and MC properties in correlation to the microstructure. The Mn-rich and Ni-rich alloys in the Ni-Mn-Sn system are chosen in view of tuning the martensite transition with optimal MC properties near room temperature. A partial Sn $\rightarrow$ Ni substitution in the Mn-rich  $\text{Ni}_{39+x}\text{Mn}_{50}\text{Sn}_{11-x}$  ( $x \leq 2$ ) series promotes the martensite phase stabilization near room temperature. In the other series  $\text{Ni}_{50}\text{Mn}_{50-x}\text{Sn}_x$  ( $12 \leq x \leq 18$ ), a small Sn $\rightarrow$  Mn substitution ( $x = 18$ ) triggers a single magnetostructural transition with functional MC properties. An advantage with the  $\text{Ni}_{39+x}\text{Mn}_{50}\text{Sn}_{11-x}$  ( $x \leq 2$ ) alloy series is that it yields inverse MC properties near room temperature.

The results of the structural, thermal, mechanical, electrical, MC, and magnetoresistance (MR) properties obtained on these two alloy series under selective experimental conditions are presented in four different Chapters 3-6. Chapter-1 gives a general introduction about the subject of ferromagnetic Heusler alloys, with the statement of the problem, review of the literature along with the motivation behind selecting this specific class of the work, and typical physical properties and applications of such alloys. Chapter 2 describes experimental details of the alloy formation and sample preparations in part of the measurements and analysis of the different properties. Chapter 3 and 4 deal with the structural transformation, Vickers microhardness, calorimetry, and consecutive magnetic transitions in the  $\text{Ni}_{39+x}\text{Mn}_{50}\text{Sn}_{11-x}$  ( $x \leq 2.0$ ) alloys. X-ray diffraction (XRD) patterns reveal a single martensite phase formed on a critical 9.0 at% Sn ( $x = 2.0$ ) content so as to exist at room temperature in a  $L1_0$  tetragonal crystal structure. At  $x = 2.0$ , MC properties arise in the martensite and ferromagnetic  $\leftrightarrow$  paramagnetic transitions concur in a narrow temperature window, with the martensite  $\leftarrow$  austenite transition up-lifted well above the room temperature  $\sim 310.5$  K. Superparamagnetic features exist below a paramagnetic regime that begins ( $\geq 250$  K) before a ferromagnetic austenite state lines-up the successive transitions. Chapter 5 describes MC and MR properties in  $\text{Ni}_{39+x}\text{Mn}_{50}\text{Sn}_{11-x}$  ( $x \leq 2.0$ ) nanocrystallites. A maximum magnetic entropy change  $\Delta S_m \sim 11.8$  J/kg-K and a large MR  $\sim (-)32\%$  have been achieved in a single phase martensite alloy ( $x = 2.0$ ) at a field change  $\Delta B = 5$  T in the transition. In Chapter-6, structural, electrical, and MC properties in a selective Ni-rich alloy  $\text{Ni}_{50}\text{Mn}_{32}\text{Sn}_{18}$  have been analyzed. The XRD pattern from the sample reveals a nanocrystalline austenite phase of a  $L2_1$  cubic crystal structure. The nanocrystallites exhibit a large  $\Delta S_m \sim 14$  J/kg-K at  $B = 10$  T, and MR  $\sim (-)15.5\%$  and giant Hall resistivity  $\sim 55$   $\mu\Omega\text{-cm}$  at the martensite transition temperature  $\sim 152$  K and a low  $\Delta B = 0.8$  T. A summary of the work with important implications achieved in this work is reproduced in the last Chapter 7 along with the future scope of the work in this series.

*Keywords:* Heusler alloy; Martensite transition; Herringbone nanostructure; Griffiths phase; Magnetocaloric and magnetoresistance properties; Magnetic entropy; Hall resistivity