

Abstract

Magnetic refrigeration based on the magnetocaloric effect (MCE) offers an energy-efficient and environmentally friendly alternative to conventional gas-based cooling technologies. However, the practical implementation of magnetocaloric materials (MCMs) is limited by challenges such as large thermal hysteresis and poor mechanical properties. The present work addresses these challenges through a systematic investigation of Mn-rich Heusler-derived alloys, specifically the Ni-Mn-Ga, Ni-Mn-Cu, and Mn-Cu-Ga ternary systems, for room-temperature magnetic refrigeration applications

An integrated approach combining CALPHAD (CALculations of PHase Diagrams)-based thermodynamic modelling and experimental alloy synthesis, was employed to identify composition windows with phase stability and magneto-structural transitions. Self-consistent thermodynamic databases were developed for the three ternary systems (Ni-Mn-Ga, Ni-Mn-Cu, and Mn-Cu-Ga), enabling accurate prediction of transition temperatures (T_C and T_M). Selected compositions were synthesized and characterized using X-ray diffraction, electron microscopy, differential scanning calorimetry, SQUID magnetometry, and mechanical test.

Mn-rich Ni-Mn-Ga alloys exhibited a giant magnetocaloric effect with a maximum isothermal entropy change of $\sim 24.5 \text{ J kg}^{-1} \text{ K}^{-1}$ at 4 T (Tesla) magnetic field and narrow thermal hysteresis near room temperature. Mn-Ni-Cu alloys showed improved mechanical ductility while maintaining a significant entropy change ($\sim 4.48 \text{ J kg}^{-1} \text{ K}^{-1}$ at 1 T), reported in this ternary system. Mn-Cu-Ga alloys demonstrated an optimal balance of moderate magnetocaloric performance, low thermal hysteresis ($\sim 1 \text{ K}$), and mechanical properties. Furthermore, Direct Ink Writing based additive manufacturing was applied to fabricate porous Ni-Mn-Ga based structures, yielding broadened working temperature range and refrigerant capacity comparable to bulk alloys.

Overall, this work establishes a unified framework for designing mechanically robust and thermodynamically stable magnetocaloric materials and provides viable pathways toward scalable magnetic refrigeration technologies.