## ABSTRACT

Magnets are essential components for domestic uses in many electric and electronic devices such as power generators, computers, medical tools, renewable energy systems, and automobiles among many more. In this investigation, a rare-earth free series of MnBi alloys has developed, which present unique features of an alternative to well-known Nd<sub>2</sub>Fe<sub>14</sub>B type rare-earth alloys for different magnet technologies. This research work presents experimental studies of structure, microstructure, and magnetic properties of pure and reinforced MnBi alloys as hybrid composites of small core-shell crystallites. The first part includes synthesis of MnBi alloys of selected Mn/Bi  $\cong$  1.0, 1.5 and 2.3 ratios, while the other part is an extension of it of developing Mn<sub>0.5</sub>Bi<sub>0.5</sub> composites, with 1-4 wt% C and 5 to 20 wt% CoFe<sub>2</sub>O<sub>4</sub>, of duly tailored properties useful for the various applications. The results obtained on the structure, microstructure, and magnetic properties under selective experimental conditions are described in four Chapters 3 to 6. Chapter-1 gives a general introduction to the subject of MnBi alloys and derivatives with the statement of problem, literature review, and motivation behind selecting the present materials, and properties and applications of such materials. Experimental details of syntheses of the alloys/composites, and measurements/analyses of their properties are described in Chapter-2. In the results and discussion, Chapter-3 deals with studies of Mn<sub>0.5</sub>Bi<sub>0.5</sub> alloy, which has grown in multiple facets of small crystallites on a local surface reaction, as on heated with a free Bi (liquid) of a precursor powder (after arc melting),  $Mn_{0.5+x}Bi_{0.5-x}-Bi$ ,  $x \le 0.05$  and  $\le 3$  at% Bi, at a critical 573 K temperature for 24 to 96 h (in Ar gas). At 300 K, a maximum coercivity (H<sub>c</sub>) = 9.850 kOe (14.435 kOe at 350 K) developed, with a net saturation magnetization ( $M_s$ ) = 72.5 emu/g, an enhanced Curie point (Tc) = 641.5 K, and an order of enhanced magnetocrystalline anisotropy (K<sub>1</sub>), in critical single domains (Dc  $\sim$  33 nm) on a due effect of a core-shell structure of small crystallites. Chapter-4 describes an antisite  $Mn \rightarrow Bi$ diffusion and its effects on an anisotropic <110> growth in a Mn-rich alloy Mn<sub>0.5+x</sub>Bi<sub>0.5-x</sub>, x  $\leq 0.2$ , on optimized anneals. A Mn  $\rightarrow$  Bi substitution (smaller atomic size) results in lower lattice parameters of a 'hexagonal crystal lattice' on anneals. A moderate  $H_c = 8.515$  kOe (12.825 kOe at 350 K) is obtained, with  $M_s \le 69.5$  emu/g, in small  $Mn_{0.6}Bi_{0.4}$  crystallites. Novel results obtained on two series of composites are described in Chapters 5 and 6, respectively. A grafted C-sp<sup>2</sup> not only tunes a huge H<sub>c</sub>, but also T<sub>C</sub> and spin reorientation (T<sub>SR</sub>) transitions. An enhanced  $H_c = 12.505$  kOe so observed, with a moderate  $M_s = 49.6$ emu/g, on 2 wt% C-sp<sup>2</sup> of a percolation threshold. Further, the CoFe<sub>2</sub>O<sub>4</sub> finely tunes combined structure and magnetic properties of the two phases, with  $M_s = 63.8 \text{ emu/g}$  at 5 K (60.5 emu/g at 300 K) at CoFe<sub>2</sub>O<sub>4</sub> up to 20 wt%. A summary of salient features achieved in this work is given in Chapter-7 along with a further scope of work in this series of MnBi type alloys and composites.

**Keywords:** Rare-earth free alloys, High-energy-density magnets, Core-shell crystallites, Hybrid nanocomposites, Exchange-coupled magnets, Magnetic properties