ABSTRACT

Intermetallic Fe_2VAl Heusler alloy has recently triggered an intense research activity to understand its unusual physical properties, such as high resistivity ($\rho \sim 10^3$ $\mu\Omega$-cm), negative temperature coefficient of resistance, moderately enhanced specific heat at low temperatures, large density of states at Fermi level and nonmagnetic behavior down to 2K. The resistivity values of this alloy are quite sensitive to the substitution of elements, which is reminiscent of doped semiconductors. Contrasting and new experimental results have appeared in the literature on this alloy and a range of microscopic mechanisms have also been proposed on the basis of theoretical and experimental investigations. However the results published on magnetic and electrical transport behavior give a rather confusing picture of the situation. In this work we report a comprehensive study of Fe_2VAl alloy and the effects of substitutions (isovalent (B,In) and extravalent (Si) elements at Al sites) on physical properties. From the structural analysis, it is confirmed that all the alloys investigated in the present study stabilize in Heusler-type $L2_1$ structure with *Fm3m* space group. A systematic analysis of magnetization and electrical transport data on Fe_2VAl alloy revealed the presence of superparamagnetic clusters with at least two sizes of moments. A cluster glass-like behavior in $Fe_2VAl_{1-x}Si_x$ alloys is found upto x=0.06 beyond which antiferromagnetic correlations start developing. On the otherhand a ferromagnetic–like order is developed with the substitution of isovalent elements *B,In* in Fe_2VAl. Further, a semiconductor-like transport and small positive values of Seebeck Coefficient (S) observed for Fe_2VAl alloy, turn to a metallic transport with large negative values of S even with a minute substitution of elements *B,In,Si.* However, substitution of isovalent *B* atom is more effective in reducing the electrical resistivity while the extravalent *Si* atom is effective in enhancement of S values. In addition, the substitution of *Si* enhances the power factor to a level larger than that of the conventional thermoelectric materials at low temperatures. Among all the alloys investigated in this work, the highest power factor value of (4.84×10^{-3} W/mK^2 at 300K) is obtained for $Fe_2VAl_{0.94}Si_{0.06}$, which appears to be a promising candidate for potential thermoelectric devices at room temperature. From these investigations, it is clear that Fe_2VAl alloy is in the vicinity of magnetic and electrical percolation threshold due to its peculiar band structure.

Keywords: Heusler alloys, $L2_1$ structure, Magnetization, electrical resistivity, Seebeck Coefficient and Power factor.

Part of this thesis work is presented in journal articles, which are listed in the list of publications.