Multifunctional materials are one of the frontline researches in the field of condensed matter physics. Multi-phase multiferroics polymer composite found their importance over single and multiphase ceramic multiferroic materials because of low saturation polarizations, brittleness causing their usefulness for device fabrication. Nano materials found to be superior to their bulk counter-part because of their novel and superior properties. Therefore, in the present dissertation, an attempt has been made to develop multiferroic-polymer nanocomposites with the introduction of spinel ferrite magnetic nano-particle into the ferroelectric PVDF polymer. The work started with the synthesis of spinel nano-ferrite particle AFe₂O₄ (A=Ni, Co, Mn) via high energy ball milling technique with 30 h, 60 h and 90 h of milling. By selecting the smallest particle size (i.e., the 90h of ball milled) powder, composites of PVDF matrix and ceramics in different ratios (i.e., (1-x) PVDF-x (AFe₂O₄)(A=Ni, Co, Mn and x=0.05,0.10,0.15)) were fabricated using a standard solution casting technique. The experimental XRD patterns were fitted with the theoretical data using MAUD Rietveld refinement technique. Based on our detailed analysis, it is found that the structure of ferrites remains invariant in Fd3m space group. The crystallite size is found decreasing with increase milling time or decreasing particle size. The XRD pattern of PVDF confirmed the existence of α-, β and γ-phases. The incorporation of spinel ferrite nano-particle into the PVDF matrix results in the increase of electro-active β-phase. The SEM micrographs of AFe₂O₄ (A=Ni, Co, Mn) reveal that the submicron grains are uniformly distributed over the surface and the grain size is found to be decreased with increase in milling time. The HRTEM micrographs of AFe₂O₄ (A=Ni, Co, Mn) with different milling time reveal the controlled particle size within 20nm which are distributed uniformly without any agglomeration. The less spherulitic microstructure of the (1-x) PVDF-x (AFe₂O₄) is observed and the size of the spherulitic decreases with increase in ferrite concentration. Dielectric properties of all the above samples were determined as a function of temperature and frequency. The dielectric constant of the spinel ferrites is strongly dependent on milling time or particle size of the materials. In case of AFe₂O₄ (A=Ni, Mn), reduction in particle size leads to the decrease in tanδ, whereas, it takes reverse trend in case of A=Co. The value of relative dielectric constant εᵣ for pure PVDF is less than that of ceramic – polymer composite. It is because of the
constrained polymer chains which obstructs the formation of electrical polarization. The impedance spectroscopy data provide the contribution of both grain and grain boundary on electrical properties of the materials. Negative Temperature coefficient of resistance (NTCR) behavior suggests the semiconducting nature. The equivalent circuit models provide an insight of structure-property relationship of the materials. Combined approach of impedance and modulus spectroscopy has given a deep insight to the contribution from grain and grain boundary on electrical properties. The temperature dependent ac conductivity shows that the value of activation energy decreases with increasing frequency. The temperature dependent ac conductivity results also indicate the dominating role of electron, polaron, ions and vacancies for the conduction process in the above materials. The conductivity behavior with respect to frequency obeys Jonscher’s power law. Study of M-H loop of spinel ferrites, NiFe$_2$O$_4$ MnFe$_2$O$_4$ with different milling time, shows ferrimagnetic ordering whereas ferromagnetic ordering is found in case CoFe$_2$O$_4$ and saturation magnetization is found to be particle size dependent. The saturation magnetization increases with increasing milling time with a slight variation at 60h of milling. The ferrite-polymer nanocomposites (1-x) PVDF-x (AFe$_2$O$_4$) (A=Ni, Co, Mn) and (x=0.05, 0.10 and 0.15) show well saturated ferroelectric loop. The saturation polarization and coercive field decrease on increasing filler concentration because of the presence of magnetostrictive phases. The composites show well saturated magnetic hysteresis, and the saturation magnetization increases with increasing filler concentration. The M-E response of the polymer composites shows an increasing trend with increase in filler concentration.

Key words: Spinel ferrite, Multiferroic, XRD, SEM, HRTEM, Dielectric, Impedance spectroscopy, Conductivity, Polarization, Magnetism