ABSTRACT

A lot of works have been devoted to the spin-polarized transport and thermal transport of the nanomaterials as evident from the already existing literature. The prime focus of the current thesis is to understand the spin-polarized electrical transport and spin-thermoelectric features of nanoscale devices and materials.

The Chapter 1 describes the general introduction of the spin-polarized electrical transport and thermal transport properties of nanomaterials. Spin polarization in a system and spin-thermoelectric features of the nanomaterials are also discussed. The background literature which is the base of the current thesis is depicted in this chapter. The prime objectives of this very thesis have also been pointed out.

In Chapter 2 the theoretical background of the magnetic exchange coupling constant and spin-polarized electrical transport are discussed in detail. The theoretical background of the thermal conductivity and spin-thermoelectric features are also outlined.

In Chapter 3 we describe the investigation about four different already synthesized meta- and para-connected diradicals to get their magnetic exchange coupling constants (*J*) through generalized valence bond (GVB) approach and also with Hartree-Fock (HF) theory with an objective to establish the superiority of GVB method over HF theory for the prediction of *J*. In doing so, at first optimization of these molecules are done at B3LYP level of density functional theory using the 6-31++g(d) basis set. With these optimized geometries, *J* values are computed at GVB method along with TZV basis set and these values are compared with those of the *J* values obtained using HF/TZV level of theory. Nonetheless, *J* values found in both the theoretical processes are also compared with the experimental findings. As a whole, the supremacy of the wavefunction based GVB method in predicting *J* is established in comparison with the HF method.

The Chapter 4 delves into the conductance property of transition metal (Ti, V, Cr, Mn, Fe, Co, Ni) encapsulated C_{20} endohedral metallofullerenes (EMFs), sandwiched between non-magnetic gold electrodes. DFT optimization of the designed systems casts the encapsulated metal at the center of the C_{20} cage irrespective of the system and spin states. Application of density functional theory coupled with non-equilibrium green function techniques on these

EMFs results variant transport characteristics for different metal encapsulation, represented through transmission spectra at zero bias and current vs. voltage (*I*-*V*) plots. Spin-polarized electrical transport has been observed for the Mn, Fe, Co and Ni encapsulated C_{20}, while no such spin polarization could be traced for Ti, V and Cr both in absence and presence of applied bias. The EMFs with Mn, Fe, Co and Ni display half-metallic behavior and significant spin-filtering efficiency, which reaches maximum value for the Fe encapsulated C_{20}. Spin-resolved electrical conductance in the Mn/Fe/Co/Ni-EMFs are explained by density of states, projected density of states at zero bias and molecular orbital analyses, which reveal the contribution of C_{20} electrons towards the down-spin-polarized transmission for Fe/Co/Ni-EMFs, while for the Mn-EMF, the metal *d*-electrons mostly participate in the up-spin-polarized transmission. Taken these observations together, the present computational study showcases the importance of encapsulating transition metal inside carbon cages in inducing spin polarization and thus provides an aid to tailor suitable molecules for spintronic applications.

In Chapter 5 we have designed a spin caloritronic device based on boron doped armchair graphene nanoribbons (B2-7AGNR). In presence of ferromagnetic graphitic-carbon nitride (*g*-C_4N_3) electrodes the spin-thermoelectric features of the device, both for ferromagnetic and aniferromagnetic states, are studied using first principle calculations. The spin polarized transmission peaks and the presence of density of states near the Fermi level indicate that the system have large spin-thermoelectric figure of merit. In addition, it is observed that the system has a large tunneling magnetoresistance due to the difference in total current between ferromagnetic and antiferromagnetic configurations. Further studies reveal that the spin component of the Seebeck coefficient of the device is much higher than the other zigzag and armchair nanoribbons. When the spin magnetic moments of the electrodes are aligned in parallel manner, spin-thermoelectric figure of merit of the system becomes significantly high. It has also been found that on decreasing temperature the efficiency of the device increases. As a whole, these results show that *g*-C_4N_3-B2-7AGNR-*g*-C_4N_3 system in ferromagnetic configuration is an efficient low temperature thermoelectric device.

A brief summary of all five chapters has been provided in Chapter 6.