CHAPTER V

CONCLUSIONS

This thesis presents a comprehensive study of the [Fe(salBzen-5-OMe)$_2$]A where A = Cl$^-$ **1**, Br$^-$**2**, I$^-$ **3**, NO$_3^-$**4**, and ClO$_4^-$ **5** and [Fe(salPren-5-OMe)$_2$]A where A = Cl$^-$ **6**, Br$^-$ **7**, I$^-$ **8**, NO$_3^-$**9**, and ClO$_4^-$ **10** complexes, focusing on the effects of the counterion (A) and the substituent group on a tridentate ligand platform (R). These complexes were successfully synthesized, and their magnetic properties thoroughly explored.

The UV-Vis spectra show two LMCT bands corresponding to the LS and HS states, supported by theoretical studies on related complexes. The crystal structures determine by X-Ray crystallography indicate that they crystallize in monoclinic P2$_1$/n or P2$_1$/c (**1**, **2**, **4** and **5**) or tetragonal P4$_3$2$_1$2 (**3**) phases. The X-ray crystal structures at room temperature reveal HS Fe(III) centers in **1**, **2** and **5**, and LS Fe(III) centers in **3** and **4**. Notably, complex **1** maintained its HS state across a wide temperature range, displaying a rare symmetry-breaking behavior unusual in Fe(III) systems. A 3D molecular network in these structures is stabilized by a variety of intermolecular interactions, including C-H···π, C-H···O, N-H···O, C-H···anion and N-H···anion. SQUID magnetometry revealed diverse SCO behaviors among the salBzen series. Complexes **1** and **2** remained in the HS state down to 10 K, while complex **5** underwent an abrupt, but incomplete SCO with a small hysteresis of 6 K, corresponding to 20% Fe switching. Complexes **3** and **4** displayed gradual SCO above 350 K, emphasizing the role of larger anions in stabilizing the LS state. For the salPren series, despite unsuccessful recrystallization attempts, magnetic studies demonstrated gradual and incomplete SCO behavior with temperature. These findings underline the critical influence of counterions and ligand substituents on spin-state stabilization in these Fe(III) complexes. The electrochemical studies of **1**–**5** and **6**–**10** highlight the impact of ligand structure and counteranions on redox behavior. The salBzen complexes show greater variation in redox potential due to stronger anion interactions within a rigid binding pocket, while the salPren complexes exhibit more consistent potentials, suggesting increased anion mobility. Ligand oxidation potentials remain largely unaffected by the anion, though the salPren series display slightly higher oxidation potentials due to their stronger electron-donating nature. The narrower peak separations observed in salPren complexes indicate faster electron transfer compared to salBzen. These results demonstrate that ligand flexibility, anion size and hydrogen bonding capability all significantly influence the redox properties of Fe(III) Schiff base complexes.

This work contributes to the understanding of structure-function relationships in SCO systems. Future studies will focus on a wider range of anions to access more abrupt and tunable SCO behaviors, with the aim of advancing applications in molecular switches and spintronic devices.