## Abstract

The present thesis has as bottom line the structural study of molecular materials. In this contribution, the capabilities of the powder X – ray diffraction technique for solving structures using conventional radiation source are emphasized. The scope and the strategies on the resolution and refinement of crystal structures are analyzed. This way, the most relevant steps in the entire process of the structure determination has been highlighted.

Such strategy developed in this work is applied to the *ab initio* structure solution and refinement of a series of coordination compounds based upon transition metal nitroprussides. As complementary techniques, the IR spectroscopy and thermogravimetric analysis are an option to introduce *a priori* information for the structural study.

Finally, the crystal structure determination and crystallographic data analysis enable to clarify the interactions governing the crystal packing for the coordination compounds. This and many other properties will be determined by both the molecular building block and the assembly metal.