Synthesis, crystalline structure and magnetic properties of a complex with benzimidazole ligand [Nibbpi$_2$]

Felipe M. Scaldini[1*], Humberto C. Garcia[1], Mario Reis[2] and Flávia C. Machado[1*]

[1]*Universidade Federal de Juiz de Fora (UFJF), Juiz de Fora, Brazil,* [2]*Universidade Federal Fluminense (UFF), Niterói, Brazil*

*e-mail: flavia.machado@ufjf.edu.br

Due to their fundamental interest and potential applications in magnetic devices, molecule-based magnetic materials have been explored intensively in the past three decades. One of the outstanding merits of this type of magnetic material is that specific magnetic traits in the molecular solids can be inserted intentionally by employing the emergent intrinsic properties of compositional metal centers and ligands[1]. Benzimidazole is a heterocyclic organic compound containing a benzene ring fused to an imidazole ring. This class of ligands shows other properties, such as in medicinal chemistry[2]. Here, we report the synthesis, crystal structure and magnetic properties of novel coordination compound named [Nibbpi$_2$], involving the ligand 2,6-bis(2-benzimidazolyl)pyridine (bbpi) and Ni(II). This compound was obtained under solvothermal conditions, where an aqueous solution containing $Ni(NO_3)_2{\cdot}6H_2O$ was mixed with a solution of DMF containing bbpi. The mixture was sealed in a 25 mL Teflon-line Parr acid digestion bomb, which was heated up to 100 °C and kept under these conditions for 90 h. Green crystals suitable for X-ray diffraction analysis were collected. The compound crystallizes in a tetragonal system presenting space group $P4_22_12$. The asymmetric unit displays the divalent nickel ion. Ni(II) center adopts an octahedral geometry and the coordination sphere consists of six nitrogen atoms, where four nitrogen atoms are from imidazole rings and two nitrogen atoms are from pyridyl rings, as seen in Figure 1. A supramolecular arrangement is achieved through N–H···N hydrogen bonds between bbpi ligands with N···N distance of 2,769 Å, (Figure 2). From the crystal structure and the plot of chiT a magnetic interaction map has been done for the complex, where the possible interaction between the copper ions was then predicted. The magnetic susceptibility was measured as a function of temperature. From the reciprocal susceptibility was observed a strong influence of the diamagnetic contribution. The value of the paramagnetic Curie temperature (-0.9K), obtained from the fitting of the reciprocal susceptibility, suggests that the complex has an antiferromagnetic overall character. The measurement of the magnetization as a function of the magnetic field suggests a local spin configuration such that the fundamental multiplet leads to a magnetic moment of 2 μB/Ni. This possible state may be associated with the transition of 17 K. From the crystal structure we can further a deeper analysis of the magnetic behavior of the compound and develop a theoretical model to adjust the magnetic data to obtain the free parameters, for instance, the exchange integral J of the isotropic Heisenberg hamiltonian, the Lande factor g and other quantities.

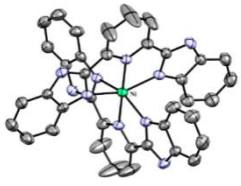

Figure 1: coordination sphere of [Nibbpi$_2$]

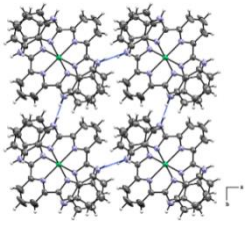

Figure 2: Supramolecular arrangement of [Nibbpi$_2$]

[1]Zhang, D., Zhuo, S., Zhang, H., Wanga, P., Jiang, J. *Dalton Trans.* **2015,** 44, 4655.
[2]Uchida M., *Chem. Pharm. Bull.* **1990**, 386, 1575.
Acknolegments: FAPEMIG, CAPES, CNPQ.