Experimental report

07/08/2019

Proposal: 5-31-2581 **Council:** 4/2018

Title: Exploring the magneto-structural correlations on Double Salt MagneticIonic Liquids: Dimim[Fe(ClxBrx-1)4] (0<x<1)

Research area: Materials

This proposal is a new proposal

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Samples: Dimim[Fe(ClxBrx-1)4] Dimim= 1,3 dimethyl imidazolium

Instrument	Requested days	Allocated days	From	To
D2B	3	3	14/09/2018	17/09/2018
D1B	3	0		

Abstract:

In recent studies, we have found that imidazolium magnetic ionic liquid based on tetrahaloferrate anions display very interesting magnetic and dynamic phenomena. Now, we want to extent the investigation to "Double Salt Magnetic Ionic Liquids" (DSMILs), which is an emerged field and, thereupon, more research is needed to understand these kind of systems in a fundamental way. For this issue, we have prepared a DSMIL solid-solution of formula Dimim[Fe(ClxBrx-1)4] x ranging from 0.95 to 0.05. To determine the unexpected magnetic behavior (with the nuclear and magnetic structure) as the function of the Br- content is the main objective of this proposal.

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Magnetic ionic liquid (MILs) based on imidazolium cation and tetrahaloferrate ion present several non-covalent interactions such as hydrogen bonds, halogen-halogen (between the nearest metal complex anions) or anion-π (between the anion and cation) interactions. These materials can show several phase transitions from room temperature and interesting magnetic phenomena at low temperatures like a three-dimensional (3D) magnetic ordering. Furthermore, a complete picture of its magnetic behavior is still missing, being extremely sensitive to temperature cooling rate effect. Therefore, using suitable rates of cooling from room temperature solid state, different ordered crystalline phases can be obtained with different magnetic behaviours. Recently, we have investigated two new MILs based on imidazolium salts and metal halides, named Dimim[FeCl4] and Dimim[FeBr4] (Dimim: 1,3-dimethylimidazolium), which are isostructural at room temperature (phase **II**) but present different nuclear and magnetic structures at low temperature (the bromide compound does not transit). Furthermore, the chloride compound presents two different structures at low temperature depending on the cooling process (called **Ia** for the quenched phase and **Ib** for the relaxed phase). In this experiment, we plan to study the crystal and magnetic structure of a series of new deuterated DSMILs (Double Salt Ionic Liquids) with 3D antiferromagnetic ordering with the chemical formula Dimim[Fe(ClxBr1-x)4], x ranging from 0.95 to 0.025, with the methyl groups of the Dimim molecule deuterated.

High resolution neutron powder diffraction patterns (D2B) with λ = 1.5942 Å were obtained for the samples with 15, 20, 35 and 50% of bromide content, at 20 K, after quenching in liquid nitrogen, and room temperature (10 scans, 25 step/scan, 500000 counts/step). This complements the previous study performed in D2B, were the samples with 2.5, 5, 10, 60 and 75% of bromide were measured. In Figure 1 we can see the patterns at low temperature for different contents of bromide, where we can see how the structure changes as we increase it. As we observed in the previous experiment, the low temperature phase for low content fits to the phase **Ia**, likewise the chloride pattern. Nevertheless, this fit worsens as we increase the content of bromide. In this experiment, we saw that the limit where this structure does not fit anymore is 50% of bromide.

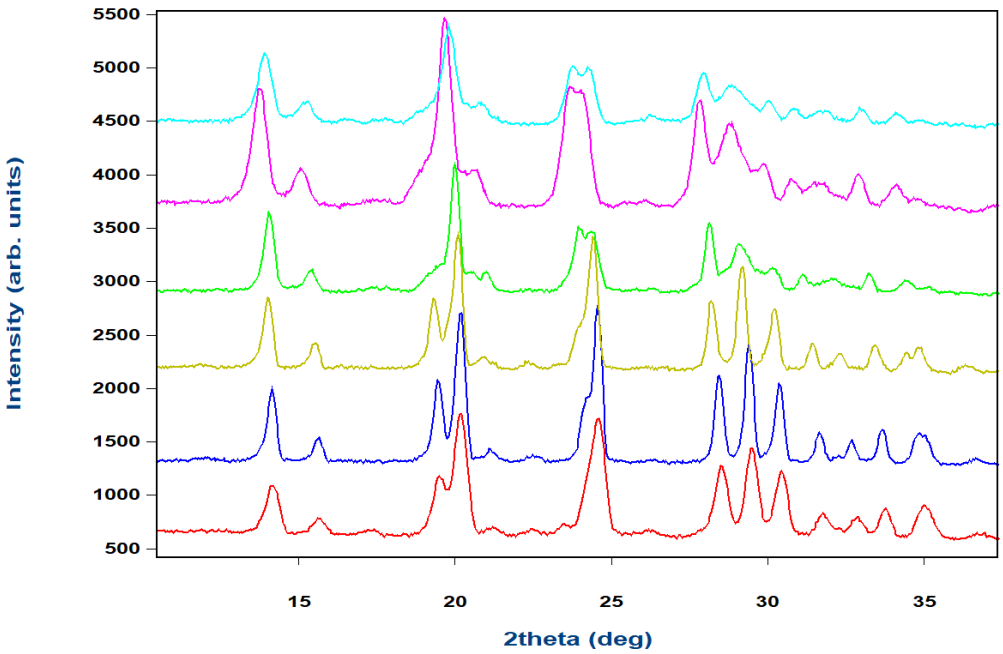

Figure 1: diffraction patterns at low T for the samples with 5 (red), 15 (blue), 35 (yellow), 50 (green), 60 (pink) and 75% (light blue) of bromide.