Experimental Report

Proposal: **5-31-2221** **Council:** 4/2012

Title: Crystal and magnetic structure of some Yb2Ti2O7 pyrochlores.

This proposal is a new proposal

Researh Area: Chemistry

Main proposer: **NUNEZ Pedro**

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Samples: Yb2Ti2O7
(Yb0.9Ca0.1)2Ti2O7-d
(Yb0.9Pr0.1)2Ti2O7-d
(Yb0.9Ca0.05Pr0.05)2 Ti2O7-d

Instrument	Req. Days	All. Days	From	To
D2B	2	2	31/10/2012	02/11/2012

Abstract:

Pyrochlores A2B2O7, a superstructure of defect fluorite (A,B)O3.5, similarly to the perovskite family of oxides, allow a wide range of cations to be introduced both to A and B sites. This remarkable tolerance to chemical substitutions makes pyrochlores a very versatile class of materials that exhibits a number of interesting physical properties. When appropriately doped, Ti-pyrochlores can be converted in good mixed or ionic conductors. However, on heating these phases show complex disordering in oxygen subllatice, which may be strongly affected by presence of dopants. Therefore we propose to study magnetic and crystal structure of several doped Ti pyrochlores in order to clear up the magnetic moment ordering at low temperature and if cation mixing between A and B sites and possible anion disordering take place at elevated temperature.

Crystal and magnetic structure of some $Yb_2Ti_2O_7$ pyrochlores.

Pyrochlore oxides are known to demonstrate a variety of technologically important properties. To give an example, ionic and mixed conductors, superconductors and geometrically frustrated magnets can be found among them. When doped, some oxide pyrochlores show remarkable disorder driven increase in ionic conductivity as the pyrochlore-fluorite phase boundary is approached. Others (e.g. Ca-doped $Gd_2Ti_2O_7$ and $Yb_2Ti_2O_7$) reach similar values of ionic conductivity even though no apparent transition to a defect fluorite structure takes place.

The objective of this experiment was to probe the disordering of several titanium pyrochlores *in situ* in the course of heating. Given the fact that these are fast-ion conductors at elevated temperature we hoped that the analysis of distribution of scattering density could reveal possible oxygen migration pathways and answer the question: are the disordering processes in the cation and anion sublattices coupled? To this end four single phase $Yb_2Ti_2O_7$-based samples were prepared and extensively characterized at ULL.

We were granted 48 hours of beam time to collect the powder diffraction patterns on the D2B instrument. The experiments were carried out at room temperature, 800°C and 1300°C. The high temperature patterns were collected using Nb cans and the standard ILL 1600°C furnace, while at room temperature the polycrystalline samples were contained in thin-walled V cans. It should be mentioned that appreciable amount of beam time was lost waiting for the furnace to cool down to about 200°C whenever the sample had to be changed, which resulted in considerable downtime on the instrument and rather poor statistics of the patterns obtained. The data collected at high temperature presented an additional difficulty: the recrystallization of Nb altered the intensity scattered at some angles dramatically. Additionally, the Nb reflections appeared split and in some cases severely overlapped with those corresponding to pyrochlore (Fig. 1).

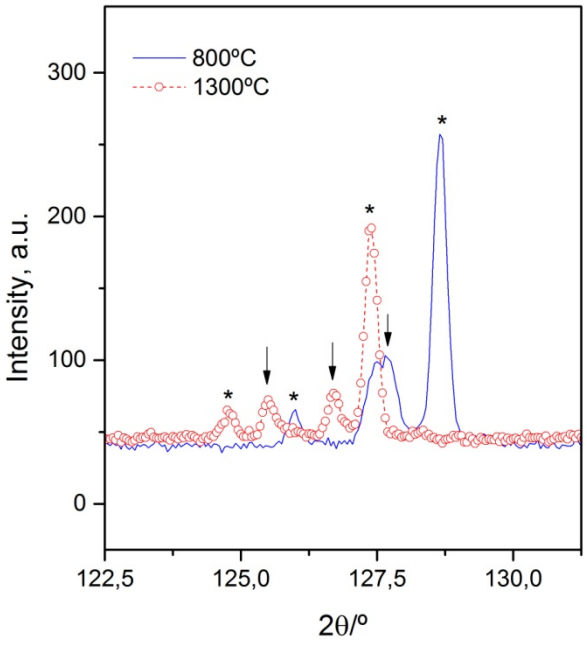

Fig. 1. Splitting of Nb reflections at high temperature. The peaks corresponding to Nb are marked with arrows and those corresponding to pyrochlore with asterisks.

The splitting increases drastically at high temperature.

Despite of the above mentioned difficulties, we have finally found out a way to treat the Nb contribution to the patterns, which allowed to fit the experimental data quite satisfactorily (Fig. 2). Currently the whole set of data is being treated by the Rietveld method.

Since no time was assigned for the determination of the magnetic structure, these measurements have not been performed.

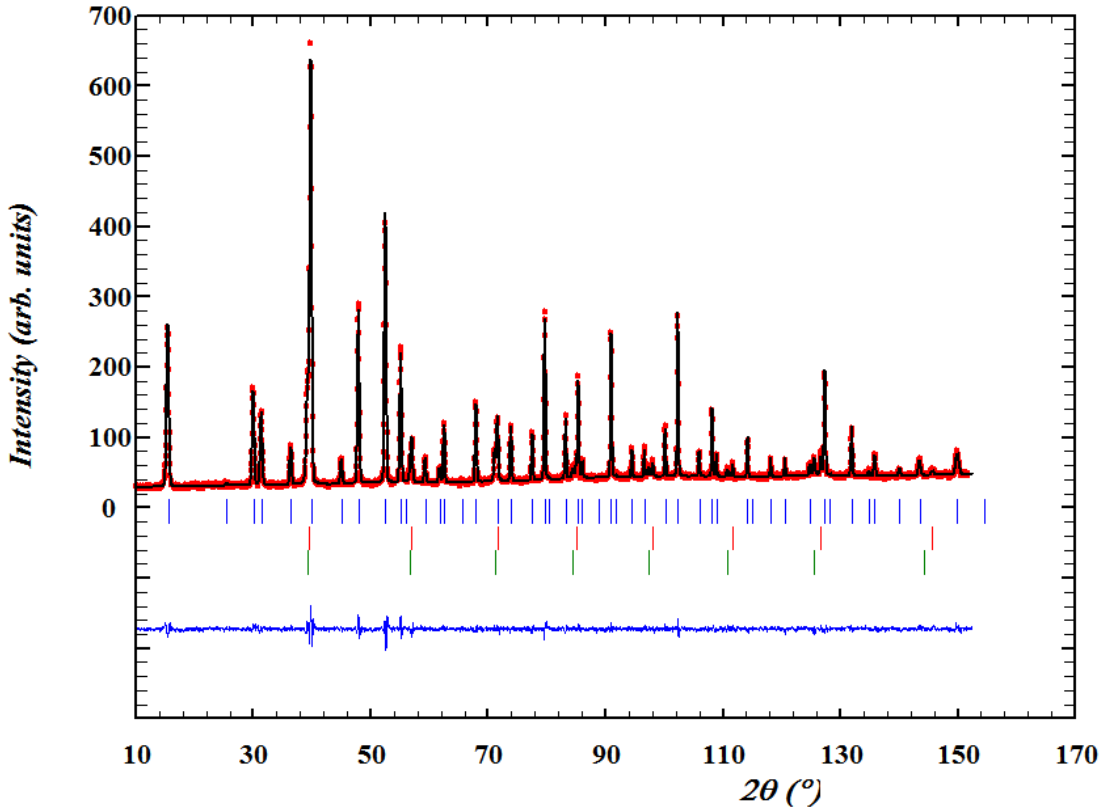

Fig. 2. Powder diffraction pattern for $Yb_2Ti_2O_7$ sample obtained at 1300°C. Experimental intensities are shown as data points and continuous line through the data points represents the fitted profile.