Experimental report

26/10/2016

Proposal:	**5-31-2427**	**Council:**	4/2015
Title:	Defining key structural and magnetic parameters for standardising magnetic Fe-oxide nanoparticles for biomedical applications		
Research area:	Physics		

This proposal is a new proposal

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Samples: Fe_3O_4 5 nm
Fe_3O_4 10nm @SiO_2
Fe_3O_4 10nm@DMSA
Fe_3O_4 80nm

Instrument	Requested days	Allocated days	From	To
D20	2	0		
D2B	4	2	25/11/2015	27/11/2015
D1B	0	4	27/11/2015	01/12/2015

Abstract:

The enormous research in magnetic nanoparticles is due to their appealing size-dependent physical properties. On top of that, promising applications are coming over. One of the more promising and at the same time regulated one, is the utilisation of nanosystems to cure and prevent illnesses. For these biomedic applications, it is mandatory to define the structural and magnetic arrangements of the nanoparticles to then select the key parameters to establish a safe standard. We have carefully selected a group of 4 freeze-dried magnetite/maghemite powders in which we have already performed an exhaustive laboratory characterisation. Our aim is to exploit the unique capabilities of neutron diffraction and define the structural and magnetic structures to compare with previous magnetisation, Mossbauer, TEM and XRD results.

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Iron-Oxide magnetic nanoparticles (MNPs) of magnetite/maghemite represent a hot topic of discussion for biomedical applications. The basic mechanisms to obtain a good performance in hyperthermia (cancer therapy) are still object of intense debate and must be connected with the magnetic structure of the cores and their shape, along with the influence of both coating and synthesis process of MNPs. With regard to this matter, it was recently prompted out the necessity of starting a standardisation route for MNPs. For this reason, we have selected a group of 7 freeze-dried magnetite/maghemite powder samples including different core-sizes, coatings and synthesis processes in which combining the neutron diffraction (ND) with other standard characterization techniques (i.e., magnetisation, Mössbauer, TEM and XRD) to study the structural and magnetic properties.

Initially, a high-resolution powder ND experiment on D2B with λ = 1.594 Å was performed at room-temperature (RT) for multicore CSIC05b (1g, Dextran, Dext. coating), "pure" Magnetite and Maghemite (2g and 1g, respectively, without coating), single-core CSIC01 (1g, SiO2) and CSIC02 (145mg, DMSA), and multicore FeraspinL (100mg, CarboxylDextran, CDex,) and BNF-Starch (1g, Starch). Samples were placed in a cylindrical vanadium sample-holder. In addition, CSIC05b and BNF-Starch were chosen as representative multicore samples to perform thermodiffractograms (5K, 100K, 200K and RT) to track the evolution of the magnetic structure from low (5K) to RT temperature, going through the blocking temperature (~100-150K). Figure 1a shows all the patterns at RT. CSIC01 and CSIC02 have the same core being, the only difference that the former has a larger coating which results in a diminishing of the diffracted pattern of CSI01. Data was collected in consecutive runs of 30 minutes, accumulating a total of 3h/sample.

Figures 1b and 1c display the patterns of CSIC05b and BNF-Starch, respectively. Samples were put into a roulette to minimise the interval of time measuring another sample in-between setting the next temperature on CSIC05b. Thus, the position of the sample along the selected temperatures was not fixed. A negative shift in the background at the 100K pattern on CSIC05b (see Fig.1b) could be consequence that all measurements in CSIC05b were not consecutive. The same procedure was followed when measuring the BNF-Starch sample. There is a shit in the position of two peaks at 200 K (marked with "* ", in green) when heating from 100K to 200K. It should be pointed out that these samples might contain sufficient amount of magnetite in their metallic core to undergo a magnetic ordering change (Verwey transition ~120K), which in turn could explain such effect.

Secondly, crystalline structure of the aforementioned set of samples was also studied from high flux neutron powder diffraction patterns on D1B λ = 2.52 Å. The time per scan was of the same order 3h/sample. We measured 5 samples: single-core CSIC02, "pure" DTU100-Magnetite and DTU0-maghemite, and the multicore samples CSIC05b and BNF-Starch (to compare data from D1B and D2B, but measuring more temperatures). Figure 2a displays the ND patterns measured at RT on D1B for all the samples, and Figure 2b shows the patterns of BNF-Starch sample at selected temperatures: 2, 50, 100, 200 and 295K.

These promising results will lead us to a better understanding of the relationship between the structure and magnetic properties of these iron-oxide MNPs.

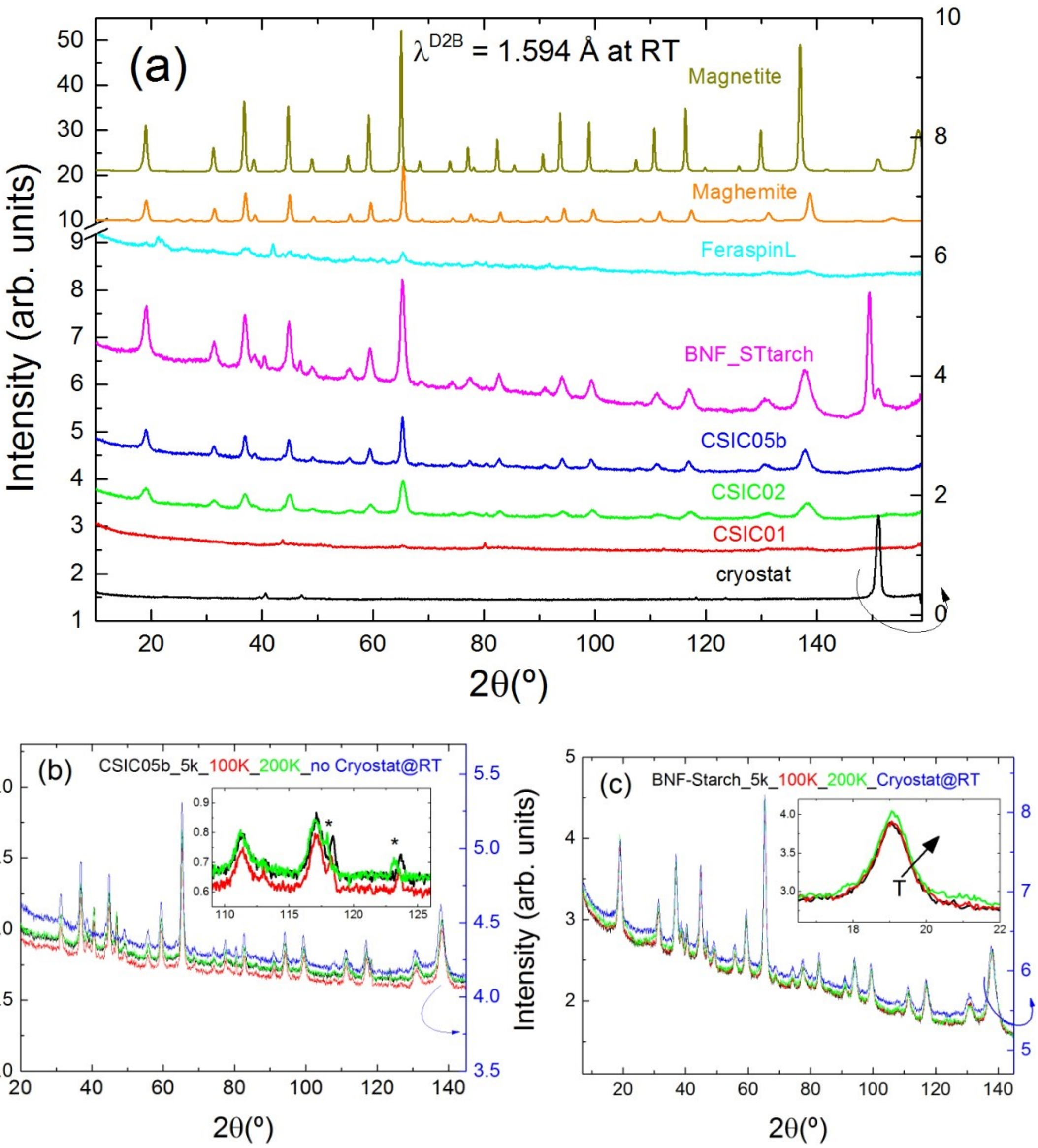

Figure 1: ND patterns measured in D2B at RT for several samples (a), and at the selected temperatures 5K-black, 100K-red, 200K-green, RT for both CSIC05b-blue (b) and BNF-Starch (c) multicore samples (b) and (c) blue RT ND patterns are resize with right axis. The insets in Figures (b) and (c) are a zoom-in.

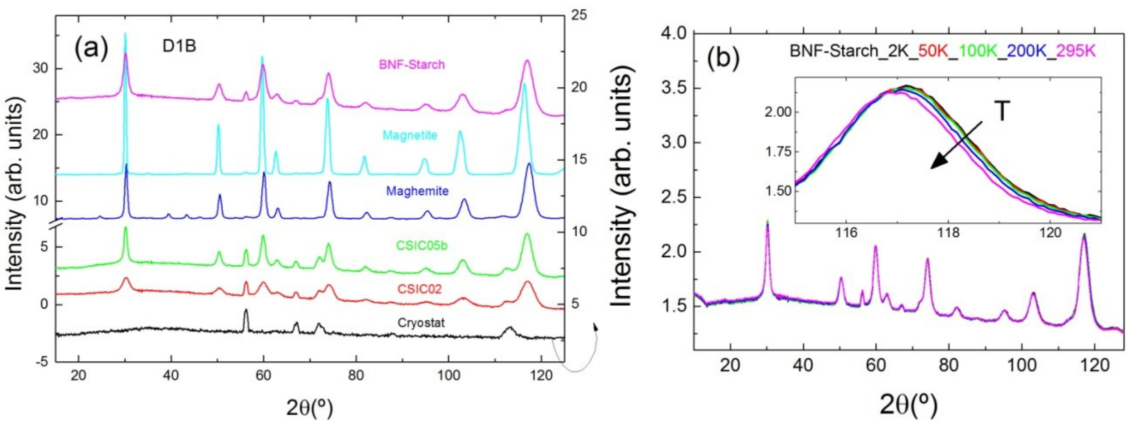

Figure 2: ND patterns measured on D1B for all the samples at RT (a), and ND patterns for BNF-Starch at selected temperatures, i.e., 2k -black, 50K-red, 100K-green, 200K-blue and 295K-magenta. The inset in (b) is a zoom-in.