ESRF	Experiment title: X-Ray Raman Scattering on magnesium intercalated fulleride polymer Mg_2C_{60}	Experiment number: HC1972
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Report:

In this experiment we wished to decipher the behavior of the recently synthesized compound Mg_2C_{60} which we had in powder and in a single crystal form.

In powder we were interested in the question: How does the C_{60} polymer network break up in high temperatures? Does the C_{60} network depolymerize, or do the individual C_{60} buckyballs break up first? The answer to this question would be interesting given the unusual thermal stability of this compound compared to its cousin Li_4C_{60}, which decomposes in much lower temperatures via de-polymerization [1]. Upon decomposition, powder diffraction pattern (for both Li_4C_{60} and Mg_2C_{60}) becomes undefined, diffraction peaks disappear, seems that the material becomes completely amorphous and thus structural information is not obtainable from x-ray diffraction.
The powder carbon K edge XRS results from annealed samples are shown in Fig. 1. We performed a thorough study on different thermally annealed samples and the changes in the spectral structure are remarkable. Preliminary analysis show that at T=700C the spectra resemble that of pure C_{60} powder while at room temperature the intensity profile, as expected, resembles a polymeric C_{60} network, when compared with existing literature [2]. The decomposition pathway is suggested to be a de-polymerization, but spectra of C K and Mg L edge highlight different features: the former displays polymer breakdown under 400C, the latter shows the same environment from RT up to 550C. These unique results, obtained thanks to the powerful IXS probe, may point in the direction of one poorly ordered phase relatively elusive to diffraction where inter-C_{60} bonding is broken and magnesium remains in intercalated form.

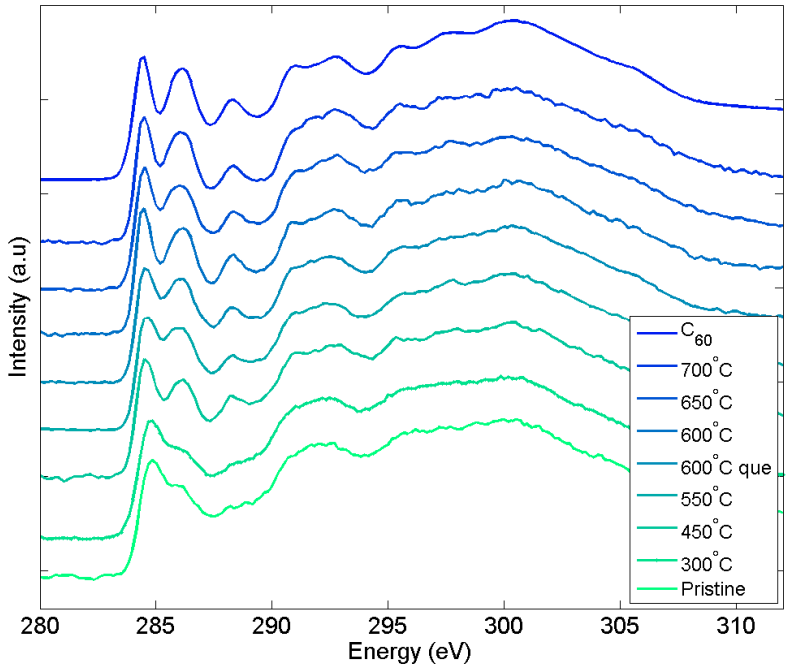

Figure 1: C K edge in different thermally annealed Mg_2C_{60} powders

Earlier measurements demonstrated that, at T~100 K, the Mg ions unfreeze from their lattice positions and the material becomes ionic conductor with a conductivity that increases with increasing temperature. Using the single crystal we were interested in the following question: How does the ionic conductivity (of Mg^{2+} ions) take place? Which pathways do the Mg ions prefer?

This challenging approach has been performed on a rare single crystal (size ~0.70* 0.50* 0.07 mm) which has been carefully aligned and oriented with q vector parallel to the (100) and to the (001) crystallographic directions. This has been achieved by modulating the energy of the incoming beam and placing suitable XRD reflections in backscattering position along the direction of the incoming beam. We measured successfully Mg $L_{2,3}$ edge at RT and at 3K in the He cryostat. Due to the small size of the crystal the measurement were unfortunately time demanding, thus only 2 lattice directions were explored. Nevertheless four measurements were completed and the anisotropy between in- and out-of-plane directions probed. The understanding of these data requires the simulation of electronic properties which will be tackled, in the forthcoming period, with the state of the art multiple scattering available algorithms.

In both parts of the experiment, data reliability was guaranteed with proper calibration and evaluation of beam damage after increasing exposure time, Compton profile was recorded in order to perform a suitable background reduction treatment.

[1] M. Riccò et al. Unusual polymerization in the Li4C60 fulleride (2005) *Phys. Rev. B,* 72, 155437. [2] Kumar et al. X-ray Raman scattering studies on C60 fullerenes and multi-walled carbon nanotubes under pressure (2007) *Diamond and Rel. Mat*. 16, 4, 1250.