ESRF	**Experiment title:** A new form of carbon – Carbon cones	**Experiment number**: HS-3330
Beamline: BM1B	**Date of experiment**: from: 04/10/2007 to: 08/10/2007	
Shifts: 6	**Local contact(s)**: Hermann Emerich	*Received at ESRF:*

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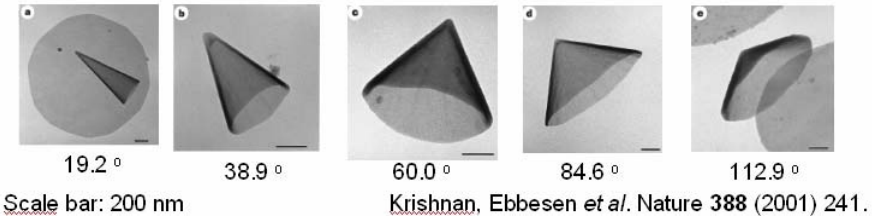
Report:

An experimental study of a new carbon material was performed in October 2007 using the powder diffraction setup at SNBL plus an addition Raman ex-situ measurement.

Carbon nanoparticles occur in five different basic forms: diamond, graphite, fullerenes, nanotubes, and the recently discovered **nanocones**. The first observation of carbon nanocones were reported by Ge and Sattler in 1994 to appear from a hot vapor phase [1], a short time after the theoretical works on this new form of carbon. All the observed cones had apex angles close to 19.2 , which is consistent with 5 pentagons at the tip of an otherwise hexagonal graphene sheet. The number of synthesized cones was small, but still remarkable, since the cone is a very seldom guest among the reaction products. Also remarkable was the subsequent finding that the geometry of the protein core shell of the HIV-virus was consistent with the topology of a carbon cone [2]. Three years later, a more efficient method to produce nanocones were found by accident. During pyrolysis of heavy oil, large quanta of open-ended cones, typically 0.5 - 1.0 micrometer long, with all the five theoretically possible apex angles surprisingly appeared [3].



The carbon cones have a perfect conical structure with symmetry fundamentally different from other known carbon materials, including nanotubes and Buckyballs. This is likely to result in different electronic-, chemical- and mechanical properties. With samples from this process as a testing ground for theoretical predictions, we are working to unveil the chemiphysical properties and nucleation mechanisms of carbon nanocones. As an example, preliminary experiments indicate significant storage capacity for hydrogen gas in carbon cones, and this application has been patented by our group [4]. The large hydrogen uptake in **powders of carbon cones** is certainly not due to physisorption or chemisorption, as is the case for other carbon materials. Possible physical mechanisms

behind this relatively high hydrogen uptake is now being investigated in our group by extensive theoretical work and computer modelling [5,6].

Very little is known so far concerning how the individual carbon layers in the cones organise with respect to each other. However, due to its particular geometrical form, for this material there is no basic "unit cell" that is repeated at fixed distances through the material (contrary to e.g. nanotubes), and the carbon nanocone powder will present itself as predominantly non-crystalline. This limits the applicability of traditional techniques for structure determination. We wanted therefore here to also prepare the ground for measuring total scattering (PDF) for these samples.

Experimental method

The experiment was done at ambient conditions in transmission mode (0.8 mm capillaries) using the high-resolution powder diffraction setup and short wavelength (0.38Å). Since attempts to separate the different elements of the carbon cone material (disks, cones with different apex angles) have not been succesful so far, the measurements were done on the as-produced carbon cone material. The short wavelength was important in order to access high wave vectors and obtain good space resolution. It also reduces unwanted effects such as absorption and multiple scattering. In order to reach wavevectors close to 20 $Å^{-1}$, scattering angles up to 60 degrees were probed. Proper background subtraction was made by taking into account empty cell scattering as well as the non-zero absorption of the sample.

In addition to the carbon nanocones samples, measurements were done under the exact same conditions on two samples that are chemically identical to, but structurally different from the first sample: [1)] carbon nanotubes, and [2)] graphite.

Results and Discussion

The diffraction data for the carbon cone raw material showed only relatively broad peaks. However, measurements on a separate sample that had been heat treated to 2700°C showed a drastically improved crystalline quality with more graphite-like crystalline peaks. These results are presented in Fig. 2. Since the raw material consists of a large amount of 180° cones (i.e. disks), a large amount of the scattering origins from the latter. By combining the powder diffraction data with with independent TEM-data, we believe now that the cones and the disks contain an amorphous outer layer that becomes more graphite-like with larger crystalline domains after heating. The addition of the amorphous layer probabably helps to stabilize the inne core of layered material. It is interesting to note that the layer packing distance was found to be 3.76Å, thus considerably higher than what is normally found in standard graphite-like material.

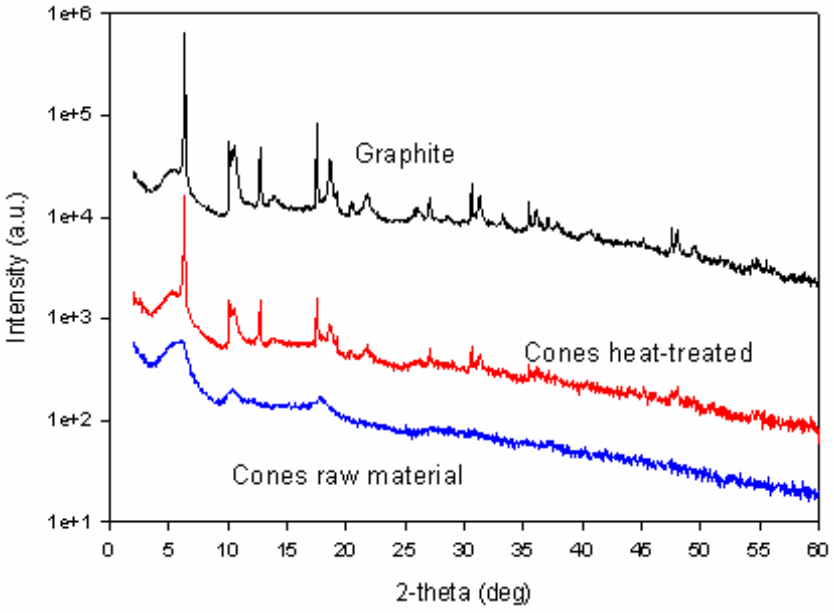


Fig. 2. High-resolution x-ray powder diffraction data (λ=0.375 Å) of the carbon cones raw material, the heat-treated (2700°C) raw material, and commercial graphite powder. The curves have been shifted vertically for clarity.

Independent electron spin resonance (ESR) measurements give a clear indication that the cone material has distinct electronic properties compared to the other probed carbon materials, and Raman data collected at SNBL ex-situ on these samples (see below) also show small shifts in the peak positions relative to the carbon reference materials that were studied. These results thus show that the carbon cones material is clearly different from other common nanocarbon systems.

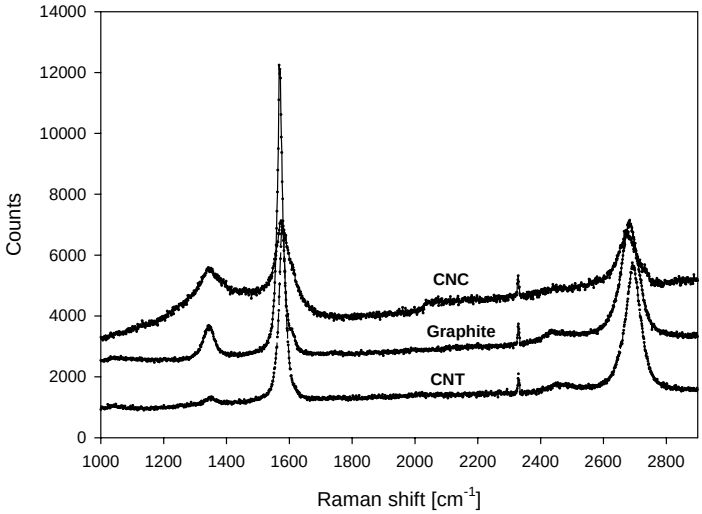


Fig.3 Raman data for carbon nanocones (CNC), carbon nanotubes (CNT) and graphite.

The study has given us important information on the construction of this new carbon material, in particular with respect to layer-layer packing and crystalline vs. amorphous regions. However, we avait further developments with respect to better separation of the different parts of the as-produced material. This has shown very difficult partly due to the fact that cones with different apex angles are chemically identical. However, work is going on here, and if better separated material becomes available, it will be an excellent candidate for e.g. total scattering studies (PDF analysis) at ESRF/SNBL.

Some of the data from the present experiment has already been published in reports to the EU-STREP project HYCONES [7].

Acknowledgement

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