

The new sample preparation line for radiocarbon measurements at the INFN Bari laboratory

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on behalf of the INFN CHNet Lilliput experiment

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Many versatile applications in the life/earth science are based on the measurement of the radiocarbon. These applications are often limited by the minimum amount of carbon that can be measured in the sample: minimum size radiocarbon samples can be affected by contamination introduced during the sample preparation. Comprehensive systematic investigations to reduce the sample size limit down to a few micrograms carbon are currently in progress in the INFN CHNet_Lilliput experiment. For such goal, a new original graphite preparation facility is being installed at the INFN laboratory of Bari (Italy). The CO₂ from the combusted sample is cryogenically purified using a simple vacuum line set-up. The produced graphite targets will be measured using the Accelerator Mass Spectrometry (AMS) at the INFN-LABEC laboratory of Florence (Italy) where since 2004, sample measurements for radiocarbon dating are performed.

Accelerator Mass Spectrometry measurements are performed on solid graphite targets at LABEC [1,2,3]. The procedure to convert a raw sample into suitable graphite involves the following stages:

- the pre-treatment process,
- the combustion of the sample,
- the graphitization process;

THE PRE-TREATMENT PROCESS

The first step of the sample pre-treatment consist of a mechanical cleaning in order to remove the outer layers of the material. Then, the samples are divided into fragments as small as possible (to maximize the available surface for the subsequent chemical attack). Subsequently, the samples are typically treated using chemical solutions depending on the fraction that has to be extracted and dated. When the possible contamination are natural, the pre-treatment is focused to essentially remove carbonates and organic soil traces, and the ABA treatment (Acid- Base-Acid) is used.

THE COMBUSTION OF THE SAMPLE

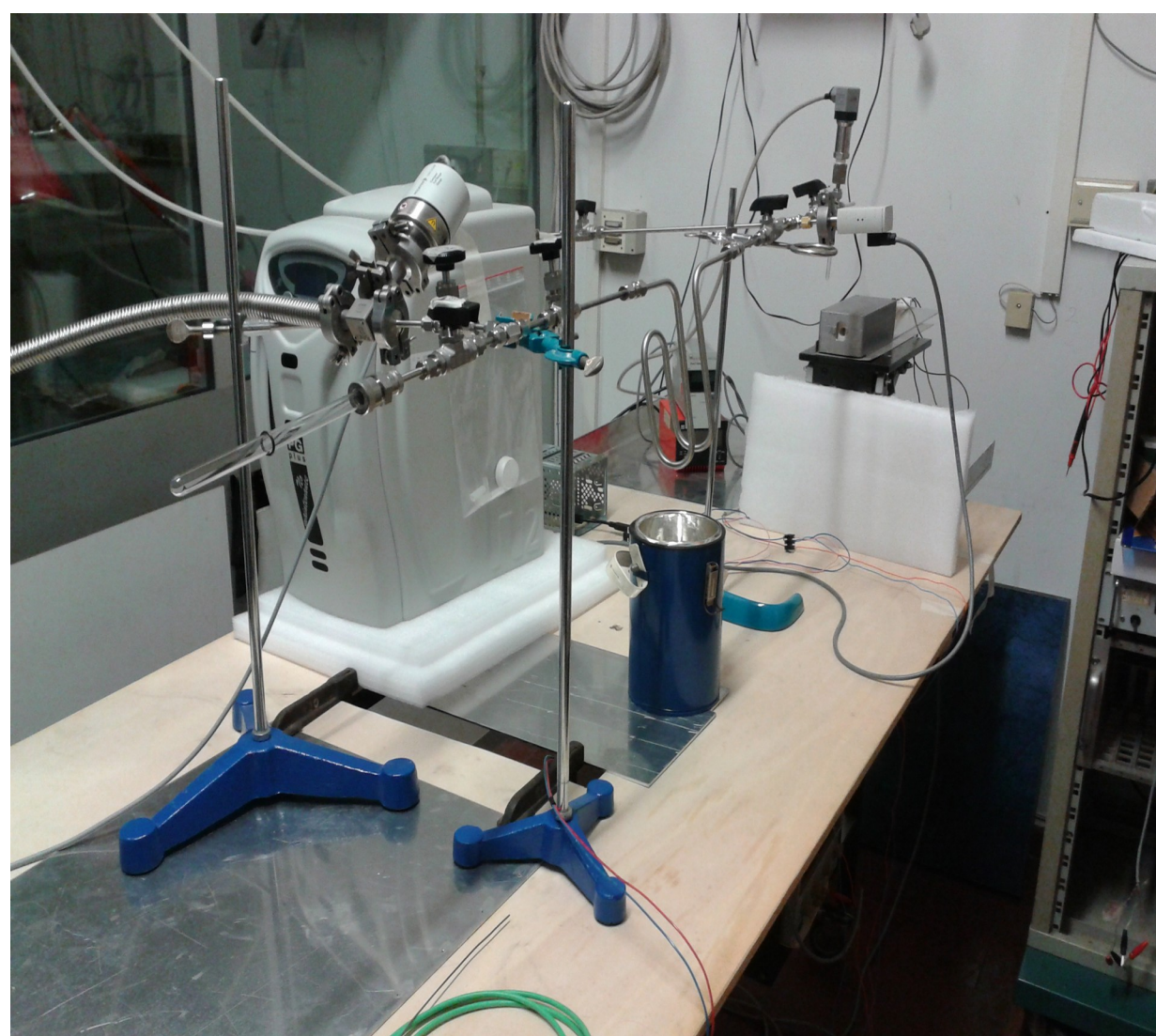
Carbon dioxide (CO₂) from organic samples is obtained by combustion the samples (in the combustion reactor, see the schematic layout) at high temperature under vacuum (~900°C), as follows:

Sample(C, H, O) + CuO + Ag → CO₂ + H₂O + non-condensable gases;

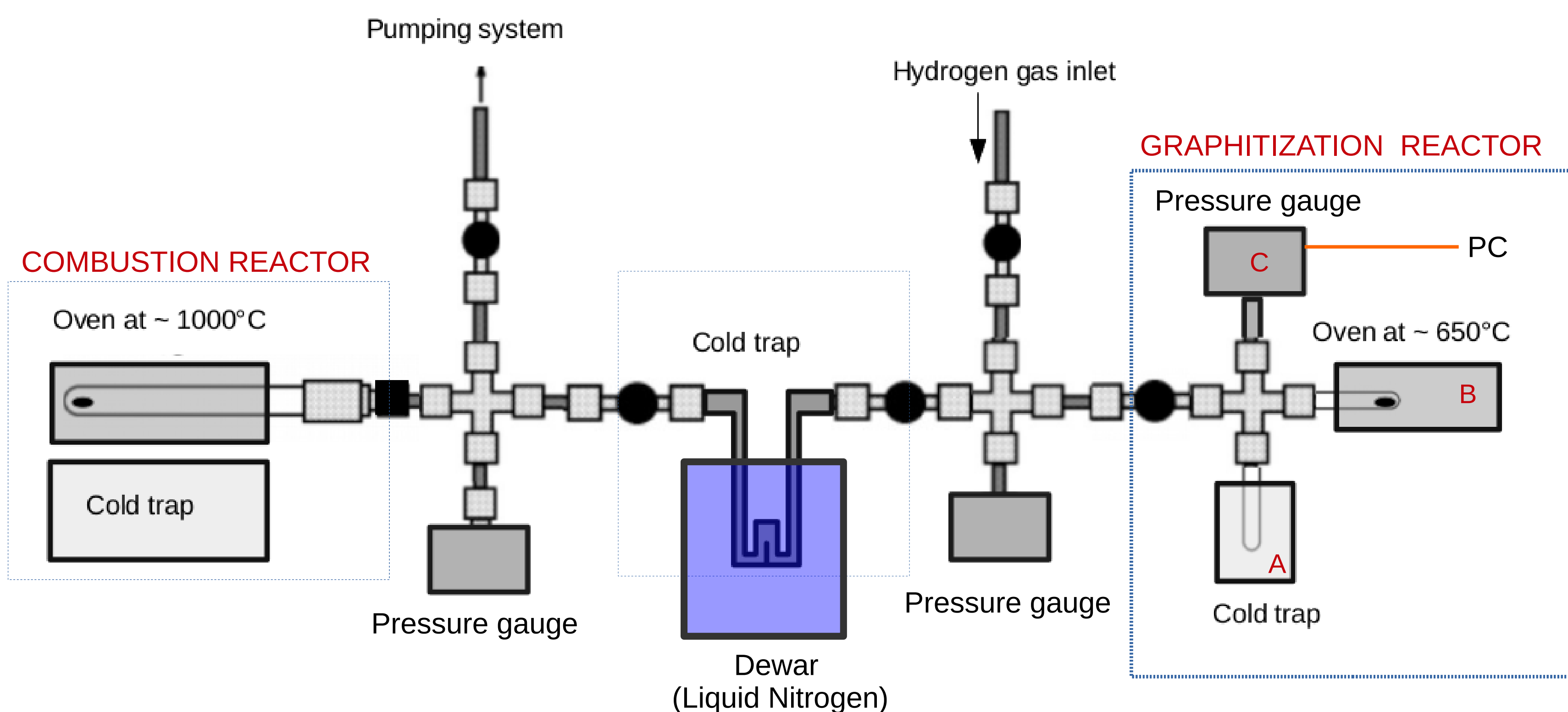
The oxygen of CuO is the primary oxygen source for the reaction. The Ag wire surface helps to remove sulphur as well as chlorine from the raw sample material.

The sample is then combusted at 900°C for 3 hours. After the combustion, the tube is inserted in a cooling device, kept at the temperature of about -25°C. The cooling device is based on the Peltier effect.

Once the water is in a solid phase, the CO₂ is collected trough the cold trap (dewar of liquid nitrogen, temperature about -196°C) and the non-condensable gases are pumped out by the pumping system. Only CO₂ will be present in the cold trap.



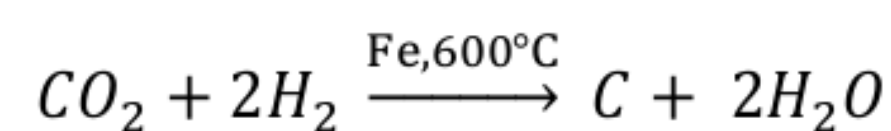
Graphitisation line installed at the INFN Bari Laboratory.



Schematic layout of the combustion and the graphitisation line.

THE GRAPHITIZATION PROCESS

The reactor is composed of two quartz tubes (length: 8mm, outer diameter: 1/4") and isolated from the line through a valve (see schematic layout). CO₂ is converted in graphite, the elemental carbon, according to the reaction:



The reaction takes place with iron powder that act as catalyst. The tube containing the catalyst is inserted in an oven (B) at the temperature of about 600°C. At the same time, the second tube is inserted in a cooling device (A) kept at the temperature of about -25°C.

The H₂O, produced during the reaction, have to be removed from the gas phase, otherwise it would progressively feed the inverse reaction. In this way, the water produced is maintained in the solid phase and the reaction can proceed until the complete reduction of all CO₂ molecules to form graphite.

The pressure inside the graphitization reactor is monitored using a pressure gauge installed in the chamber (C). An Arduino board (Mega 2560) allows the monitoring online of the pressure. Fig.1, the H₂ pressure as a function of the time inside the graphitization reactor (volume ~ 1 cm³) is shown: no leaks are present.

The graphitization process takes about two hours and half: when the chamber pressure reaches a minimum and constant value, the reaction is complete and a graphite (+iron) powder is obtained. Fig. 2, a typical trend is shown. The mixture of graphite and iron powder is pressed in an aluminium support to be inserted in the AMS source for radiocarbon measurement.

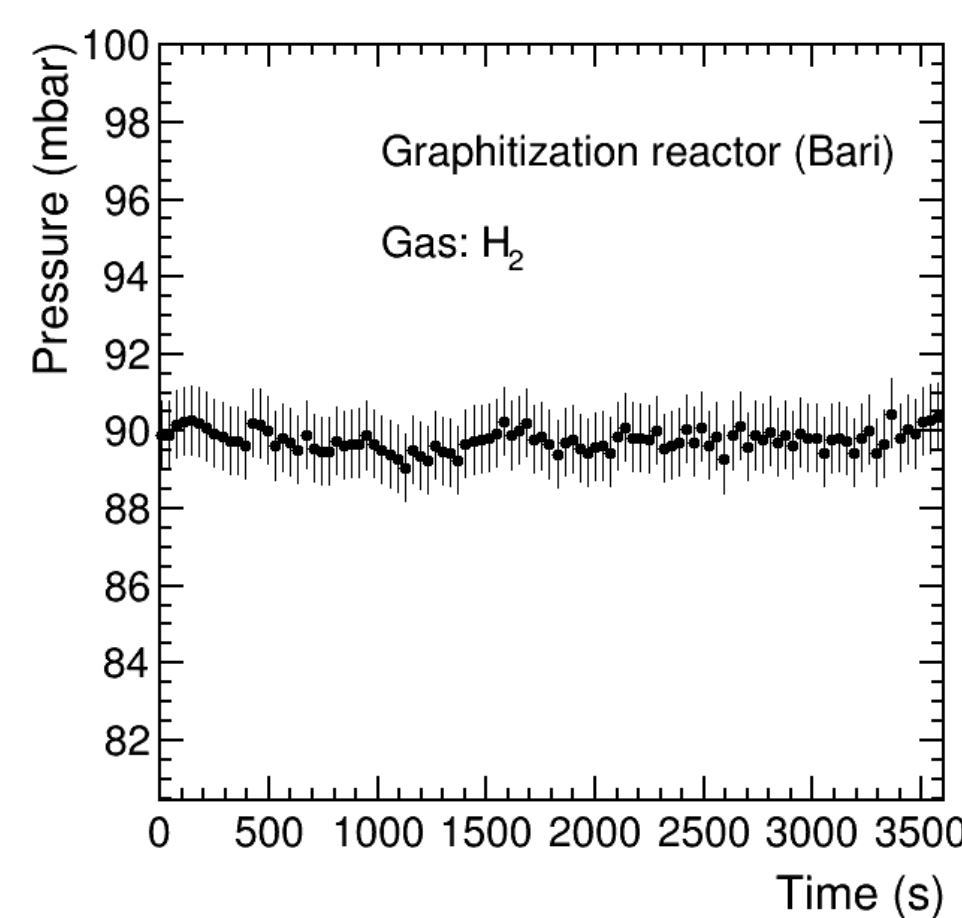


Fig. 1

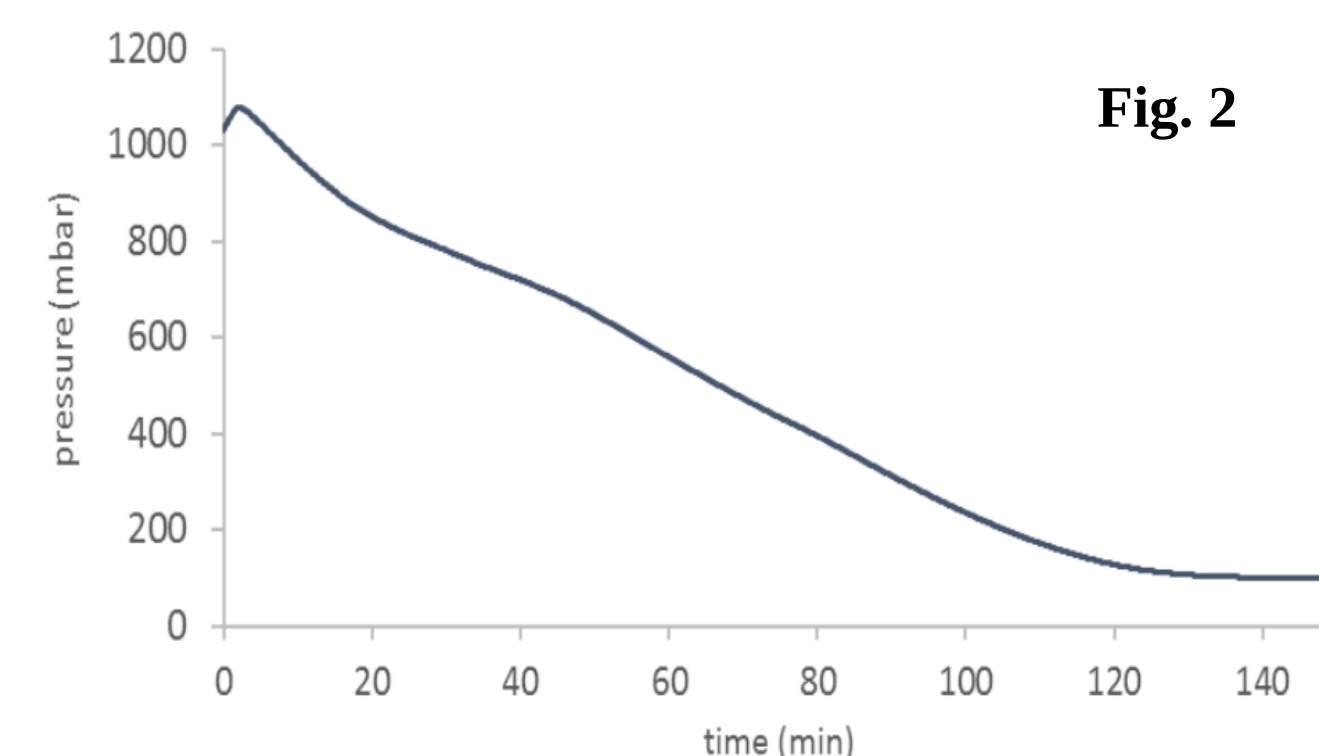


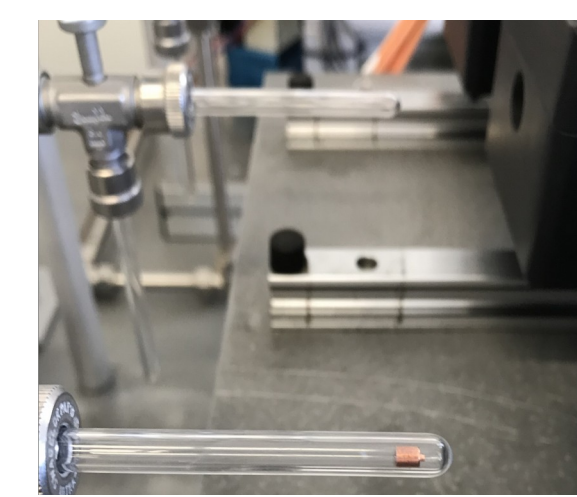
Fig. 2

TOWARD SMALLER SAMPLES

When dealing with carbon samples that are smaller and smaller, one of the most critical aspect during preparation is probability of loosing some of the collected materials while passing from one stage to the following. To overcome this possible problem, we are developing a system to directly graphitise samples on already pressed iron powder. The forst sample (mass ~ 1/3 typical sizes) were succesfully prepared and measured at LABEC.



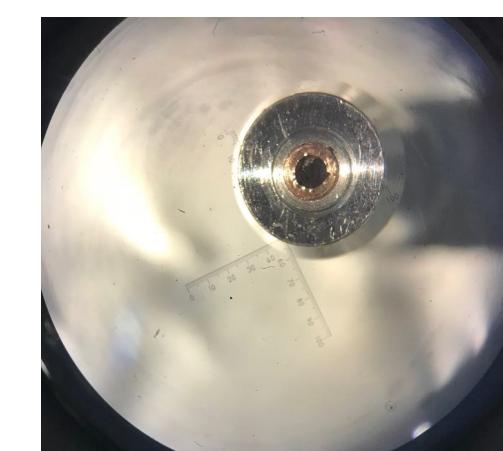
A)



B)

A) Press iron powder into small copper holders

B) Insert the copper holder into the graphitization reactor



C)

C) Insert the copper holder (iron + graphite) into the typical accelerator source target holder

REFERENCE

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