A study on diamond grains contents in primitive meteorites and ureilites using micro-IBIL and micro-PIXE techniques.

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I. INTRODUCTION

In this research we tried to obtain new information about diamond genesis in primitive meteorites and ureilites using ionoluminescence and PIXE facilities at Legnaro microbeam.

Primitive meteorites contain a few parts per million of pristine interstellar grains that provide information on nuclear and chemical processes in stars. Their interstellar origin is proven by the highly anomalous isotopic ratios, varying more than 1000-fold for elements like C and N. Microdiamonds or nanodiamonds, with medium size of few nm, are the most abundant and still poorly known. They contain anomalous noble gases which show the signature of the r- and p-processes and thus are apparently derived from supernovae or at least show some connection with them. As far as the formation of microcrystalline diamonds is concerned, several hypothesis are considered. It has been noticed that the carbon stars atmosphere is very similar to the atmosphere present in CVD deposition apparatuses (temperatures between 900 and 1500 °C, H/C ratio of 400, C/O ratio of 1.5) and therefore, apart from $^{13}C/^{12}C$ ratio consideration, it could be deduced that carbon stars are also connected with microdiamonds formation. Photolysis (or also radiolysis) of hydrocarbons could be possible, with a consequent formation of diamonds by homogeneous nucleation in the gas phase. Interstellar shocks could be also a possible mechanism, because at the very high velocities of colliding carbon grains the instantaneous temperature and pressure could be enough to allow for diamond formation as in HP-HT growth method. As a matter of fact, it has been prove by using explosives that it is possible to obtain from a carbon charge small nanodiamonds with controlled dimensions not far from those reported for interstellar grains.

Ureilites are a group of achondrites unique in containing relatively large amounts of C occurring as diamond, graphite or lonsdaleite. Diamond genesis in ureilites can be related to three different possible processes that was proposed in the past: a HPHT process like in Earth or in parent bodies of lunar mass or greater, a shock conversion of graphite during a bodies impact or a CVD process (disequilibrium condensate from a gas phase) in the solar nebula. From recent works it seem that the second hypothesis is the prevalent. However, there are not still conclusive evidence for the origin of diamond in ureilites, despite the presence of pronounced crystallographic orientation in ureilites, shocked silicates and compressed graphite.

IBIL (Ion Beam Induced Luminescence) is a very powerful technique for material investigation and particularly for diamond analysis. Using ionoluminescence the presence of nitrogen in diamond (the main impurity in natural diamonds) can be detected even if it appears at ppm concentrations [1]. Nitrogen aggregation in diamond is a kinetic phenomenon in which the degree of aggregation depends on the mantle residence time of diamond, its nitrogen content, the temperature history of the specimen and the genesis (HPHT or CVD). It is known that nitrogen, at small percentages, speeds up diamond growth, and it stabilise better CVD diamond because it helps in vacancies compensation. Hence, the peak position and intensity (that is high for relevant concentration of nitrogen) in luminescence depend from the diamond type (i.e. Ib, IaA, etc.) and can be used to identify not only the nitrogen quantity or its aggregation state but also the origin of diamond.

II. EXPERIMENTAL SET-UP

Ionoluminescence data were obtained by a new version of the IBIL apparatus [2] described in in another LNL report in this year [Manfredotti et al., Ionoluminescence in CVD diamond]. The only novelty in the apparatus for this work is the nitrogen temperature cooling system we mounted recently on sample holder inside the analysis chamber. This up-grade was necessary to avoid peaks enlargement and suppression due to the high room temperature.

III. STATUS OF THE ART

Due to the higher concentration of relatively great diamond grains (few micrometers of diameter), we started analysing ureilites.
We were able to identify quickly carbon region using micro-PIXE (the dark region in fig. 1). No information about phase distribution (graphite or diamond) came from PIXE technique, so we used ionoluminescence analysis in monochromatic mode (monochromator centered at 2.88 eV, the A-band emission) to identify diamond grains inside carbon regions. In fact, this wavelength emission is a characteristic of almost all types of diamonds (stronger in type IIa diamond because the presence of other peaks due to nitrogen or other impurity can quench it) and so it can be used to identify this carbon phase. The origin of A-band emission is still debated even if the radiative recombination at dislocation is the main explanation. In fig. 2 is reported the ionoluminescence map (2.88 eV photon emission) of the same region of fig. 1. We can observe the position of diamond grains inside graphite phase.

In fig. 3 is shown a characteristic spectrum: other than the blue A-band we can observe the presence of a damaging peak (at about 2.42 eV) observed in all types of diamond after irradiation [1].

Mapping samples at different wavelength we found the presence of a second photon emission band centered at about 2 eV.

This band, we will investigate in future, is not related to all grains of diamond and so it is possible there are two type diamond species. This band may be related to N-V centres in low nitrogen contents diamond. In fig. 4 is shown the IBIL map at 2 eV.

In conclusion we were able to identify and characterise diamond grains in carbon meteoritic inclusion by means IBIL and PIXE techniques. Diamond founded are mainly type IIa diamond due to the low nitrogen concentration (presence of A-band and absence of N3 centre or other features involving high nitrogen concentration).
