Abstract:

The silicon route is one of the approaches which have been proposed for the redefinition of the kilogram in terms of the Avogadro constant. In this framework, the elemental characterization of the adopted ultra-pure $^{28}$Si enriched materials is of fundamental importance to avoid biased results. The aim of this research is the development of an analytical method involving Neutron Activation to investigate with the lowest possible uncertainty, $^{30}$Si mole fraction and quantify impurities present within the crystal.

Scientific community made great efforts in the last decades to move units of International System of measurement from artifact to fundamental constants of nature. The redefinition of unit of mass is involved in this activity. In this framework the knowledge of Avogadro constant by counting the atoms present in a quasi perfect silicon sphere highly enriched of the isotope 28 plays a key role.

In order to quantify Avogadro constant with requested relative uncertainty (< 2 x 10^-8) these silicon materials have to be of well known isotopic composition and ideally free from crystal defects. For this reason the contribution to the relative uncertainty due to isotopic composition and defects have to be below 10^-9 level, in order to use these crystals for a useful evaluation of Avogadro constant.

The aim of this project of research is to apply Instrumental Neutron Activation Analysis (INAA) for characterization of these silicon crystals. This analytical technique allows to reach low detection limits for several elements and is not destructive for the sample.

Three areas of investigation:

$^{30}$Si mole fraction

The $^{30}$Si mole fraction has been measured by several National Metrology Institutes (NMI) using mass spectrometry. In one case discrepancies have been highlighted.

For this project, a relative Instrumental Neutron Activation Analysis method was performed. A natural silicon crystal of mass and shape similar to the sample to be measured was used as standard reference. The mole fraction of $^{30}$Si was quantified by counting gamma-delayed emissions with a HPGe detector. Activation was performed by the neutron reaction $^{30}$Si(n,$\gamma$)$^{31}$Si. The method allows to reach low detection limits for several elements and is not destructive for the sample.

A preliminary quantification of vacancy-related defects was carried out using a sample of natural silicon crystal.

The sample was filled by Cu at high temperature (750 °C) for 3 h and slowly cooled to precipitate CuSi$_3$ within the void defects following the method as suggested by Spaepen.

5 following cycles of annealing at lower temperature (450 °C) for 2 h were performed to out-diffuse the interstitial Cu. Relative INAA was exploited to quantify Cu amount through the reaction $^{60}$Cu($n,\gamma$)$^{61}$Cu and counting $^{64}$Cu (12.7 h half-life) gamma emission at 1345.77 keV.

The result of the experiment showed a Cu concentration plateau corresponding to Cu trapped within the sample. From its quantification, an upper limit for the density of vacancies is estimated.