Hydrogen Analysis of Minerals from the Earth Mantle ◇

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Studies in recent years have shown that several nominally anhydrous minerals (NAM) in the Earth's mantle store low but significant concentrations of hydrogen in the order of few to few tens ppm hydrogen atoms , that are structurally mainly bound as hydroxyl ions. These low amounts are of specific interest as they provide a huge mantle-water storage mechanism and in total a globally important reservoir of hydrogen [1].

The main analytical methods for studies of NAMs has been Fourier–transform Infrared Spectroscopy (FTIR) and Secondary Ion Mass Spectroscopy (SIMS). FTIR allows to distinguish the phases of the incorporated hydrogen (if not H_2) and yields information about orientation of the H-bonds. Both, FTIR and SIMS have high sensitivity corresponding to around few tens of ppm(at) hydrogen. However, for both methods the signal yield depends on structure and other factors and hence independent analytical methods are required for quantitative statements. Additionally, hydrogen mapping at microscopic resolution is essential, because the sample structure is often inhomogeneous, the sample size of natural as well as synthesized samples are limited to few hundreds of μm and samples may contain structural defects like fluid inclusions on a microscopic scale.

Therefore, proton–proton(pp)–scattering applied to a nuclear microprobe is the method of choice for hydrogen analysis when high sensitivity or microscopic resolution or both is required. It is intrinsically self–calibrating, i.e. does not need reference material for quantification, and gives a sensitivity of better than 0.1 ppm(at) at sub–micron resolution [2]. Pp–scattering has already been used for quantitative studies of thin NAM samples recently [3]. However, beam energy limits the maximum sample thickness as both protons scattered off from a hydrogen atom (at an angle of 90° to each other) have to pass the sample in transmission geometry. Proton energies larger 20 MeV as provided by the Munich tandem accelerator are necessary to investigate NAMs that are at best to be polished down to $50-100\mu$m. Next to the beam energy requirement, it is the detector system that has to detect the full remaining energy of both scattered protons in order to provide a depth information of the hydrogen signals. The large area Si-strip detectors that are used for coincidence analysis with good enough time and energy resolution are limited to a thickness of 1 mm. Hence, a maximum incident proton energy of only 17 MeV is the limit in the case of pp–scattering with our old setup [2] when the detector surface is placed perpendicular to the beam axis.

Our new detector concept is shown in Fig. 1 on the right where the coincidence strip detectors are placed in parallel to the beam axis resulting in 2 mm effective Si thickness in the case of a proton scattered at 30° (high energy of the proton pair). With this it will be able to use up to 28 MeV protons and investigate NAMs up to around 300 μm, that are easily polishable or even to be embedded onto a glass substrate if the dimensions are to small to mount directly.

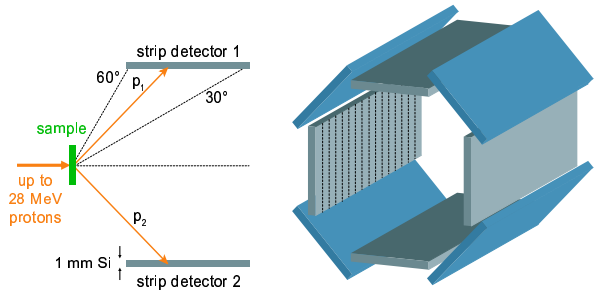

Fig. 1: New detector concept based on parallel detector arrangement of 1 mm thick silicon strip detectors.

For first experiments we used a preliminary setup as shown in Fig. 1 on the left with only two 6 cm × 4 cm large 1 mm thick detectors with 7 strips each. Using an unfocused proton beam with 25 MeV at the Q3D target chamber we analysed several natural NAM samples of collaborators from the GeoForschungszentrum Potzdam [4] and the Universität Erlangen [5]. Even with this simple setup and a solid angle of 2×92 msr we were able to reach a detection limit as good as 2−3 ppm(at) hydrogen. Future plans are to install the new detector setup at the microprobe SNAKE and start systematic studies of NAMs with μm resolution in order to clear doubts and inconsistencies of the existing IR calibration models.

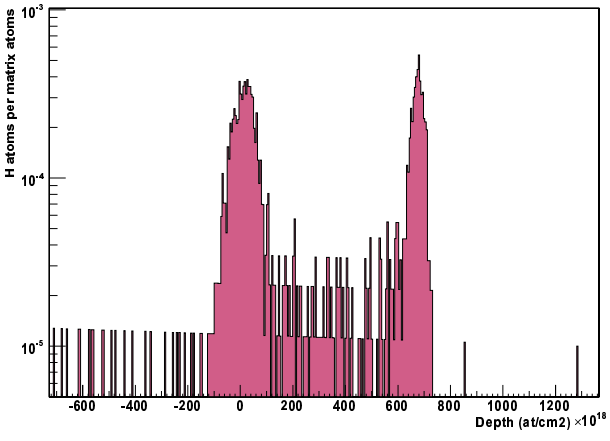

Fig. 2: Depth profile of NAM from Nigeria. The hydrogen content in the bulk area corresponds to (11.6 ± 2.6) ppm(at). The peaks are caused by 3.7 and 2.6 H–atoms/cm^2 surface contamination.

References

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