The Lund Nuclear Microprobe Facility

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Development of a system for determination of the $^{13}C/^{12}C$ isotopic ratio with high spatial resolution

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Introduction

Assessing the biogenicity of potential bacterial fossils is not an easy task, as recently shown by the controversy concerning "Earth's oldest fossils" [1]. A thorough investigation of the geological setting and the morphology and chemical properties of the potential microfossils is essential in order to assess them as *bona fide* microfossils.

In this context, stable carbon isotopes play a certain role in the interpretation of biological activity, particularly when the fossil record is studied. It is known that stable carbon isotope fractionation is associated with the fixation of CO_2 in autotrophic organisms [2]. The fractionation is dominated by the ^{13}C depletion associated with the uptake of CO_2, but also with enzymatic discrimination in the synthesis of biomass from inorganic carbon [2]. The isotopic composition of preserved organic carbon in potential microfossils may therefore be useful in the assessment of the biogenicity, as well as in the interpretation of the biological activity.

Idea

- Use the ^{13}C α-resonance at 2.75 MeV for precision measurements of the $^{13}C/^{12}C$-ratio.
- A useful geological and paleontological tool if the precision is be better than about 2 ‰.
- Solid angle as large as possible will be necessary for the required precision.
- Kinematic effects on the energy resolution have to be minimised.

Experimental tests

- The experiments have been performed at the Lund Nuclear Microprobe facility [3].
- The beam energy used was 2,75 MeV with a typical α-current of 0.5 to 1 nA.
- He-ions was extracted and transported through the standard microprobe line into the microprobe chamber. Beam size was typical 10 µm.

Test 1
Pure carbon standard (^{13}C = -25.6±0.2 vs. PDB).
A typical spectrum from such a standard is shown in figure 1 top.
The ^{13}C resonance peak is clearly seen and well separated from the ^{12}C continuum.
Yield from this RBS technique, with optimised solid angle and beam conditions, has the correct order of magnitude

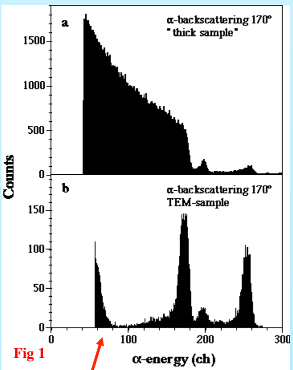


Fig 1

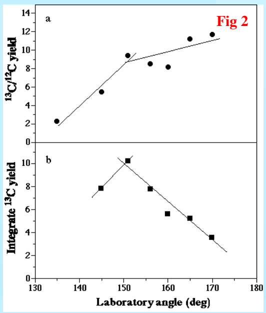


Fig 2

Test 2
Good statistics implies a detector solid angle as large as possible.
Angular yield distribution was measured, shown in figure 2. **The angular interval to be used should cover at least 150-180°.**

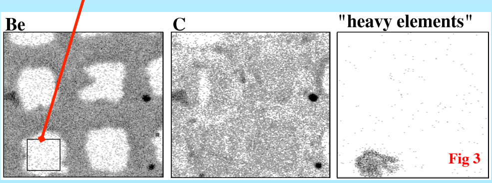
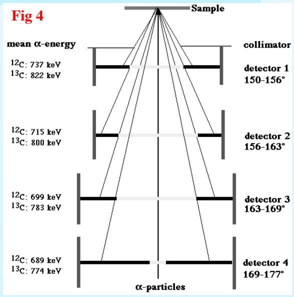


Fig 3

Test 3
In the final test experiments the sample was sectioned and placed on grids.
Both a Cu-grid and a Be-grid were tested.In figure 1b and figure 3 the advantage of this technique is obvious.
Thin samples (TEM-sectioned) should be used for the analysis.

Fig 4



System design and test

Design
Energy difference between recoils from ^{13}C and ^{12}C in the angular interval 150-180 degrees is about 85 keV.
Energy variation for the recoils from ^{13}C is about 100 keV.
Typical resolution of an annular detector of 25 keV.
Energy overlap between recoils from ^{12}C and ^{13}C.
Necessary to segment the angular interval.
Four detectors - the energy difference between the highest recoil energy from ^{12}C and the lowest from ^{13}C can be chosen to be larger than 60 keV.
The final design of a four annular surface barrier detectors system is shown in figure 4.
The solid angle covered is about one steradian (sr).

Yield
In the test experiment on the Be-grid a solid angle of about 40 msr was used and with count rate of about 0,2 count/nC.
Extrapolating, a count rate of 5/nC could be expected for the detector system.
With 2‰ statistical precision, at least 250 000 counts are necessary.
New system - total charge of 50 µC is needed, i.e. 12-13 hours with a two nA alpha beam.

Test of new system
First results from a test run with the new detector system is shown in figure 5.
The four spectra each corresponds to data from one annular surface barrier detector and all show separated ^{13}C peaks.
The run was performed with a 200 pA beam current in six hours, i.e. about 4 µC.
The total counts in the ^{13}C peaks are about 13000 and this verify the estimates given above about current and time needed for a useful analysis.

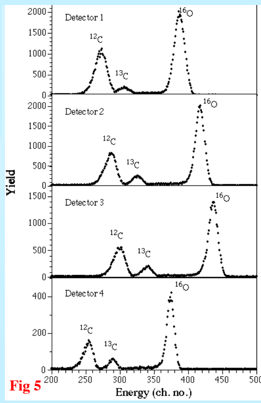


Fig 5

Future development

- The data has to be extracted with well-defined fitting routines.
- Different ways to normalise the data with high enough precision have to be investigated. Variation in thickness of the sample will introduce effects especially in the ^{12}C yield.
- The energy stability of the method has to be investigated. Does the accelerator energy need to be recorded for each event?
- Does different sample thickness influence the ^{13}C yield or is there some tolerance when selecting beam energy?

Summary

- **The possibility to use the 13C α-resonance at 2.75 MeV has been experimentally investigated.**
- **A multi-detector system based on annular detectors has been designed and tested.**
- **The possible integrated yield fulfils the statistical requirement for measuring the 13C/12C ratio with precision better than 2‰.**
- **Further development is needed for evaluation and normalisation.**

References

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