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Preliminary results of oxygen isotope ratio measurement with a particle-gamma coincidence method

Maciek Borysiuk *, Per Kristiansson, Linus Ros, Nassem S. Abdel, Mikael Elfman, Charlotta Nilsson, Jan Pallon

Lund University, Department of Physics, Division of Nuclear Physics, Sölvegatan 27, 223 62 Lund, Sweden

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ABSTRACT

The possibility to study variations in the oxygen isotopic ratio with photon tagged nuclear reaction analysis (pNRA) is evaluated in the current work. The experiment described in the article was performed at Lund Ion Beam Analysis Facility (LIBAF) with a 2 MeV deuteron beam.

Isotopic fractionation of light elements such as carbon, oxygen and nitrogen is the basis of many analytical tools in hydrology, geology, paleobiology and paleogeology. IBA methods provide one possible tool for measurement of isotopic content. During this experimental run we focused on measurement of the oxygen isotopic ratio. The measurement of stable isotopes of oxygen has a number of applications; the particular one driving the current investigation belongs to the field of astrogeology and specifically evaluation of fossil extraterrestrial material.

There are three stable isotopes of oxygen: ^{16}O, ^{17}O and ^{18}O. We procured samples highly enriched with all three isotopes. Isotopes ^{16}O and ^{18}O were easily detected in the enriched samples, but no significant signal from ^{17}O was detected in the same samples. The measured yield was too low to detect ^{18}O in a sample with natural abundances of oxygen isotopes, at least in the current experimental setup, but the spectral line from the reaction with ^{16}O was clearly visible.

1. Introduction

Chromite is a refractory mineral with the chemical formula $FeCr_2O_4$. It can be found in both terrestrial and extraterrestrial geological material. This mineral, when found on Earth, serves as the main source of chrome. The extraterrestrial variety can be found in certain types of chondritic meteoroids, which constitute a large fraction of meteoritic material falling onto Earth [1]. A meteorite will be subjected to the same chemical and physical processes, the sum of which is known in geology as diagenesis, as all the terrestrial rocks. These processes tend to severely alter the composition of minerals. After a sufficiently long time only the outline of the now fossil meteorite remains; most of the matrix has been completely transformed [1]. The fact that makes chromite interesting is its refractory nature. Refractory in geological terms means that this mineral can survive entry into Earth's atmosphere and all subsequent chemical interaction that the meteorite is subjected to, due to its exposure to Earth's atmosphere and due to diagenesis [1].

This high resistance to the elements is a valuable property, as it means that the material will preserve its ordinal composition and is compatible with IBA methods. The ratio of stable oxygen isotopes in the many small bodies found in the solar system is believed to reflect the abundances of oxygen isotopes in the presolar nebula. This is because many of the meteoroids remained largely unaltered since their formation early in the life of our solar system.

There are three stable oxygen isotopes. They are present within Earth's crust in the average proportions of 99.75% for ^{16}O, 0.03% for ^{17}O and 0.2% for ^{18}O. Their exact abundances in actual samples will vary but the fact is that the ratio of $^{18}O/^{16}O$ will fall along a straight line when plotted against the ratio of oxygen $^{17}O/^{16}O$ in all minerals found on Earth. It has been found that extraterrestrial material falls outside of this line, meaning minerals throughout our solar system have oxygen abundances deviating from the values normally found on Earth [2]. A typical oxygen-three isotope diagram can be seen in Fig. 1. An accurate measurement of all three isotopes provides a method of differentiating terrestrial from extraterrestrial material and furthermore can be used to assigning a meteorite found on earth to a specific group of meteorites [1,2].

In our earlier work, we have shown that the reaction $p + {}^{18}O$ at 0.8 MeV [3] can be used for mapping of oxygen distribution in

* Corresponding author.
E-mail address: maciek.borysiuk@pixe.lth.se (M. Borysiuk).

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0168-583X/

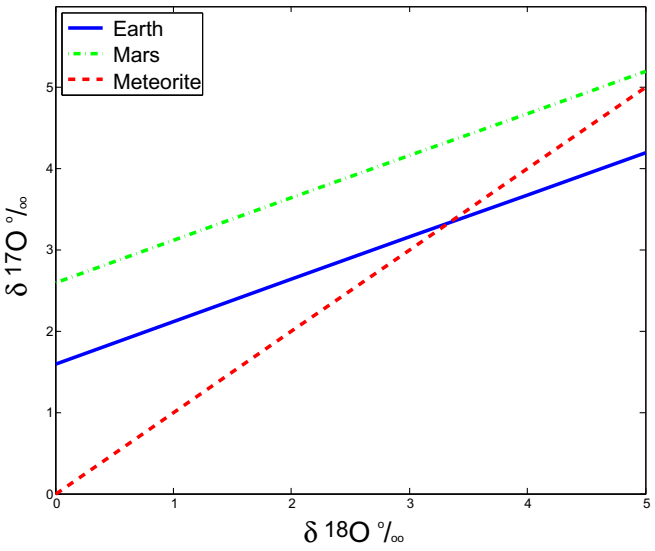

Fig. 1. A schematic example of an oxygen three-isotope diagram. Fractionation line for Earth, Mars and one of the possible ones for extraterrestrial material have been sketched in the figure.

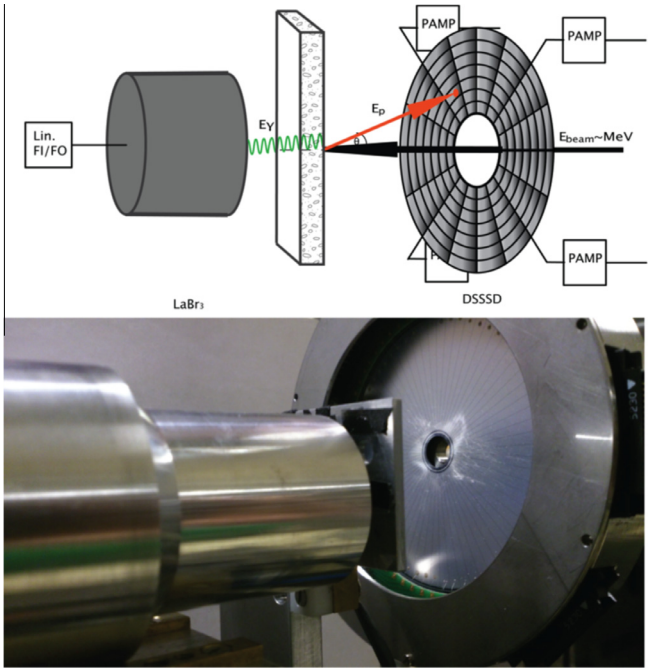

Fig. 2. A schematic description of the detector setup (above). The DSSSD is placed in the backscattering geometry so that it covers 2.5 sr of solid angle. Beam enters from the right in the figure. Gamma detector, in this case a fast scintillator ($LaBr_3(Ce)$) is placed in a close geometry after the sample so that it covers 3.6 sr of solid angle. The signal from the DSSSD is shaped using standard NIM electronics. Output of the PMT is split (which is suggested by a FI/FO box) and delayed to produce both energy and timing information necessary to capture the coincidences. Reactions generating both a gamma and a particle can be easily detected using the setup. A photograph of the detector setup in the experimental chamber is shown in the image (below).

grains of chromite extracted from fossil meteorites. The aim of this work is to find a technique suitable for mapping all three stable isotopes, simultaneously if possible, in order to be able to measure the variation of oxygen isotope ratios relative to a standard material. There are a number of known resonances for 1–2 MeV deuterons which can be useful for ^{16}O and ^{18}O determination [4], sadly the ^{17}O cross section is largely unexplored. The challenge is to find reactions with high enough cross-sections to give high accuracy and low detection limit.

2. Method

Most commonly, practitioners of Ion Beam Analysis look for light elements in samples by detecting the gamma quantum or a particle emitted in a nuclear reaction, which means proton induced gamma ray emission (PIGE) and nuclear reaction analysis (NRA) methods, respectively. Those two techniques have already been investigated as a source of isotopic information in various solid samples, see for instance review articles [5,6]. The conclusion of the previous work was that in most practical situations, especially in the microprobe setup, the reaction yields will be too low to perform accurate measurements without risking damage to the sample. It was suggested in [6] that there are several steps that can make isotopic quantification in a microprobe a viable solution. Among them are increasing the detection efficiency, by for instance increasing the solid-angle of the detector, and suppressing or discriminating background reactions. The current experiment utilizes both of those steps.

Photon tagged nuclear reaction analysis (pNRA) is a particle-gamma coincidence method first presented in [7] and extensively discussed in [8]. There it was proposed that the utility of nuclear reactions, which are routinely used for spectroscopy of light elements in various materials [9], could be increased if the focus was placed on reactions emitting both a charged particle and a gamma quantum. The simultaneous detection of both emitted radiation types with good resolution and high efficiency, provides a method of background suppression and separation of interesting reactions from each other.

A schematic representation of the situation in the reaction chamber as well as a photograph can be seen in Fig. 2. Both detectors used are brought close to the sample to maximize solid angle. In the current experiment the charged particle detector is located in the backward hemisphere, placed at a distance of 3 cm from the sample, covering 2.5 sr. The gamma detector is placed at a distance of less than 0.5 cm from the sample in the forward hemisphere, covering a solid angle of 3.6 sr. The particle detector used was a double sided silicon strip detector (DSSSD) with 64 radial strips and 32 concentric rings with an active inner diameter of 14 mm and an active outer diameter of 85 mm, described in detail in [10]. The gamma detector is a Cerium doped Lanthanum Bromide $LaBr_3(Ce)$ scintillator crystal [11], coupled with a PMT, especially chosen for its combined good timing and energy resolution [12]. The large solid angle of the setup and the segmented nature of the DSSSD mean that the large fraction of the yield produced in the various reactions in the sample is immediately available for analysis.

Our detectors are connected in a coincidence mode using standard NIM electronics. The shapers connected to the DSSSD provide timing, energy and trigger outputs. The PMT has only one output signal which is split to provide us with energy, timing and trigger for the system. The signals are delayed and the triggers from both detectors are used to build a coincidence gate. The setup used in the current experiment was described in greater detail in [12].

For the purpose of this experiment, two batches of isotopically enriched Al_2O_3 powder were purchased from Icon Isotopes [13] and then pressed into pellets at Lund Ion Beam Analysis Facility (LIBAF). This was compared to the Al_2O_3 powder with natural abundances.

In the current investigation the focus was shifted from the more commonly used proton induced reactions towards the deuteron induced reactions. For this purpose a 2 MeV D^+ beam was extracted and focused at the sub-micron beam line [14] at LIBAF. The beam current was varied between 0.1 and 1 nA depending on the sample