ACCELERATOR MASS SPECTROMETRY AT THE LUND PELLETRON

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Accelerator mass spectrometry (AMS) is a highly sensitive method for counting atoms, both radionuclides and stable nuclides. The main advantages of AMS compared to conventional radiometric methods are the use of smaller samples (mg size) and shorter measuring times (less than one hour). In this report some current applications of the AMS technique at the Lund Pelletron accelerator are presented.

1 Introduction

One of the applications of nuclear physics techniques which has been greatly beneficial and vital to other fields of scientific endeavour is accelerator mass spectrometry (AMS). AMS has not only allowed refinements in the technique of carbon dating in the fields of archaeology and quaternary geology, but has also been demonstrated to be useful *e.g.* for: tracing ^{26}Al in living systems; tackling hydrogeological problems by the use of ^{10}Be and ^{36}Cl; and following transport of ^{129}I and transuranes from nuclear facilities.

In AMS, rare isotopes from a sample material placed in the ion source of an electrostatic tandem accelerator are measured by counting individual atoms with nuclear detection techniques after acceleration to energies in the MeV range. AMS evolved from nuclear physics laboratories, and several tandem accelerators originally installed during the 60's and 70's (and therefore of insufficient energy for today's nuclear physics experiments) have been rebuilt to be used for AMS.

2 Applications at the Lund AMS facility

2.1 Radiocarbon dating

Natural ^{14}C ($T_{1/2}$ = 5730 years) can be used in numerous applications, of which radiocarbon dating, with the possibility for dating back to about 50 000

years, is among the best-known and most practised applications. Radiocarbon dating is based on the property that the activity concentration of ^{14}C in dead tissues can be used to calculate the time that has elapsed since death occurred. This is possible since all living organisms contain nearly the same proportion of radioactive carbon in their carbon stores and since, upon the death of the organism, the carbon which survives decomposition continuously loses ^{14}C by its radioactive decay.

The ^{14}C dating program at the Lund AMS facility is a collaboration between the Department of Nuclear Physics and the Department of Quaternary Geology. About 200 ^{14}C datings are performed per year at the AMS laboratory. The most common types of material used for AMS dating in Lund are charcoal, wood, bone, peat, marine shells and terrestrial macrofossils.

2.2 Studies of ^{14}C releases from nuclear power plants

^{14}C is one of the radionuclides which are produced to different degrees by neutron-induced reactions in all types of nuclear reactors. Part of the ^{14}C created in reactors is continuously released as air-borne effluents in various chemical forms (such as CO_2, CO and hydrocarbons) through the ventilation system of the power plant to the surrounding environment during normal reactor operation.

An extensive investigation of the ^{14}C releases from some Swedish nuclear power plants has been performed at the Lund AMS facility. Air emitted from the stacks was collected continuously over two-week periods and the ^{14}C contents in the samples were analysed. The chemical form of the effluents has also been studied and the ^{14}C concentration in willow leaves in the vicinity of a power plant has been measured. Measurements of the ^{14}C activity concentration in annual tree rings and air around some Swedish nuclear power plants have recently been presented [1].

As an example, the ^{14}C content of air samples collected in the down-wind direction at various distances from the nuclear power plant of Barsebäck (south west Scania, 2 BWRs each 600 MW) is shown in Fig. 1.

2.3 Biomedical applications of ^{14}C

^{14}C is used as a radioactive tracer in clinical nuclear medicine and it is used in various contexts in medical research and when testing new pharmaceuticals on volunteers. One way of carrying out these studies is to use "breath tests". The ^{14}C-labelled compound is ingested and metabolised, resulting in the end-product carbon dioxide, which is exhaled and easily collected for measurement. The decay of the radionuclide is usually measured by gas flow counters or liquid

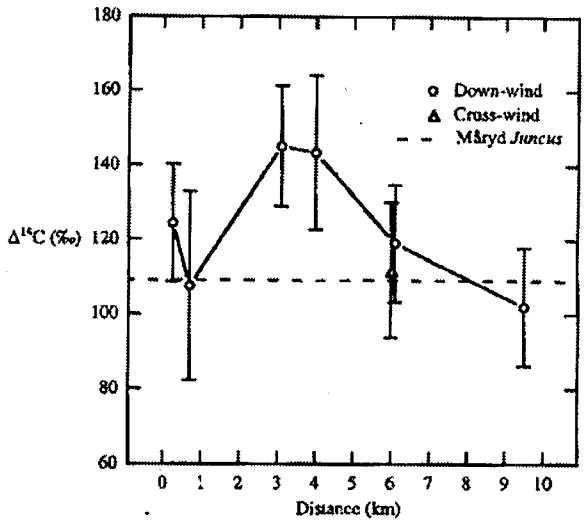

Figure 1: The ^{14}C content of air samples collected at various down-wind distances and at one cross-wind site from the Barsebäck nuclear power plant. The 1996 year value of *Juncus* from Måryd (a clean air site 30 km east of Barsebäck) is also indicated. The ^{14}C content is given as the excess of ^{14}C (in $^{0}/_{00}$) compared to the 1950 year standard.

scintillators and the activity of the sample reveals the degree of, for example, fat malabsorption.

The AMS technique has in Lund been used to study the long-term retention of ^{14}C after a fat-malabsorption test (using ^{14}C-labelled triolein) by analysis of expired air. Studies are also being performed on the long-term retention of ^{14}C after a ^{14}C-urea test, which is used to demonstrate abnormal activity of gastrointestinal bacteria. The use of the AMS technique makes it possible to follow the turnover of the radionuclide administered for extensive periods and also to decrease the activity administered [2].

As an example of our biomedical investigations, the ^{14}C specific activity in expired air at various times after administration of ^{14}C-triolein is shown in Fig. 2. The dip in the curve after 6 days (A) came after the volunteer had eaten three meals within a short time and this result motivated some controlled fasting periods (32 hours long) when the volunteer was allowed only to drink water. Such fasting periods started on days marked B to E. When the fast began the exhalation of $^{14}CO_2$ increased, and then it decreased to its starting value when the person began to eat again. During the fast, stored body fat is used to a higher degree and since most of the administered ^{14}C is still stored in the body, the ^{14}C concentration in the expired air will increase significantly.

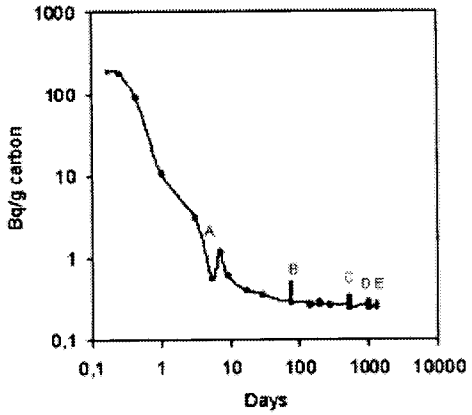

Figure 2: The ^{14}C specific activity (in Bq per gram carbon) in expired air at various times after administration of 74 kBq ^{14}C-triolein to a 94 kg male. The background was determined from 8 samples taken before the ^{14}C-triolein administration and found to be 0.258 Bq/g_{carbon}. A-E are described in the text.

2.4 Applications of ^{14}C in food chemistry

An important topic in food chemistry technology is the interaction between foods and packaging materials, the composition, quality or physical properties of the food and/or package can be altered. Several methods, each one with advantages and disadvantages, have been and are being used in studies of interactions between foods and packaging materials. In Lund the AMS technique has been introduced into food chemistry with the aim of demonstrating that AMS can be a complementary and suitable tool for investigations of the absorption of flavours in plastic packaging materials[3].

2.5 Detection of ^{59}Ni in stainless steel

In nuclear waste management, ^{59}Ni is a most important radioisotope, since it is produced by neutron activation in the stainless steel shielding surrounding the fuel. The total activity concentration of ^{59}Ni, as well as of other radionuclides, has to be established in preparation for final disposal. Because ^{59}Ni decays only via electron capture and has a very long half-life (7.6×10^4 years), it is quite difficult to measure the radiation emitted in its radioactive decay. The atom counting approach of AMS would in this case be advantageous. However, for small tandem accelerators, such as the Pelletron in Lund, the common energy or energy loss detection techniques are not able to distinguish atomic isobars for heavy elements such as Ni. One way to eliminate this problem is

to combine AMS with the detection of characteristic projectile X-rays. After analysis in the AMS system, the ions are stopped in a suitable target and it is possible to identify the ions by atomic number and thereby separate the isobars. Some promising preliminary measurements using the projectile X-ray detection technique have been performed [4]. Recently, the method has been further refined and a method to extract nickel chemically from stainless steel will soon be presented as will a determination of the detection limit of ^{59}Ni at the Lund AMS-system.

3 Conclusions and future plans

Only a few of all the possible long-lived isotopes have at the moment been used for AMS. An area which will grow in the future is biomedical studies. In the near future, large hospitals will install dedicated accelerators for ^{14}C analysis with a high sample throughput. Small quantities of, for example, isotope enriched or radioactive marked chemical substances will be used. In this way the radiation dose to the patients will be more or less eliminated.

In Lund, further development will include installation of a recirculating pump in the high-voltage terminal stripper system. In this way an enhanced gasstripper efficiency is expected. The installation of a new injector, including a spherical electrostatic analyser and a 90° magnet will give a better mass resolution, especially for heavy atoms. This development will be carried out mainly for biomedical purposes.

References

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