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Measurement of Long-lived Nuclides with AMS and Its Applications at CIAE*

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Abstract: The Accelerator Mass Spectrometry facility at the China Institute of Atomic Energy was established in 1989. The measurement methods for interesting nuclides such as ³⁶Cl, ¹⁰Be, ¹²⁹I, ²⁶Al, ⁷⁹Se and ⁴¹Ca etc. have been established and studies on their applications have been carried out. The facility and some interesting applications are briefly introduced.

Key words: long-lived nuclide; accelerator mass spectrometry; application

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1 Introduction

Accelerator mass spectrometry (AMS) is an analytical technique to detect the long-lived radionuclides that cannot be practically analyzed with decay counting or conventional mass spectrometry. AMS has been used for the analysis of ¹⁴C, ¹⁰Be, ³⁶Cl and other cosmogenic radionuclides in archaeology, geology and environmental science since the first AMS ¹⁴C measurement was carried out on the FN tandem accelerator at McMaster University in Canada 27 years ago. Recently, AMS applications have also expanded to biomedicine, radiopharmaceutical studies, semiconductors, mineral exploration and other branches of science and technology, where it is of interest to determine very low concentrations of specific nuclides. At present, AMS applications are contributing to a vast array of high priority issues related to global climate, environmental pollution, public health and international safeguards of nuclear materials.

There has been sixteen years since the China Institute of Atomic Energy (CIAE)-AMS became operational in 1989. The HI-13 tandem accelerator in CIAE is a multi-user facility. Only a small part of beam time can be used for AMS. It is impossible to measure large number of samples and to carry out studies over wider range of applications by using the HI-13 tandem AMS system. Therefore, we focused our studies on the development of measurement methods and several interesting applications utilizing the long-lived radionuclides ³⁶Cl, ¹⁰Be, ¹²⁹I, ²⁶Al, ⁷⁹Se and ⁴¹Ca etc.

2 CIAE-AMS System

2.1 Facility layout

A typical AMS facility includes the ion injection system, the accelerator, the analyzing components and detectors. Fig. 1 shows the experimental layout of the CIAE-AMS facility. The major components from the sputter ion source to the detector

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are;

(1) Ion source. The model MC-SNICS (Multi-Cathode Source of Negative Ions by Cesium Sputtering) cesium sputter ion source manufactured by NEC is used. The sputter target holder is made of pure copper or aluminum. There is a tar-

get wheel in the sputter source; forty holders with the samples can be positioned on the wheel at one time. The samples on the wheel can be rapidly changed one by one without purging the source vacuum so as to keep constant operation conditions for a group of samples to be measured.

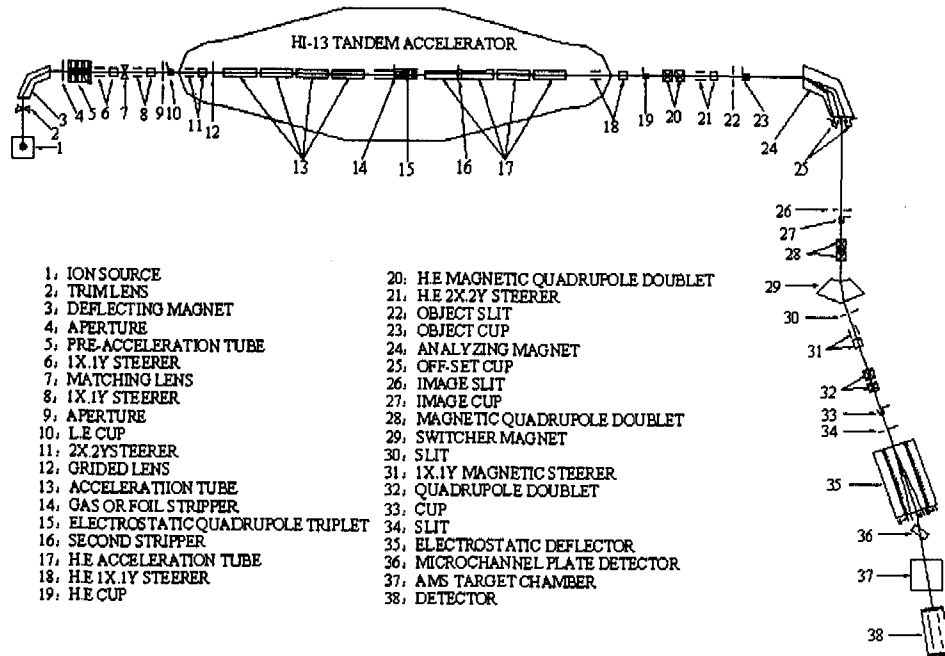


Fig. 1 The Schematic drawing of CIAE-AMS system.

(2) Injection magnet. A double focusing 90 degree deflecting magnet with a curvature radius of 36 cm is used. This magnet has a mass resolution $M/\Delta M$ of more than 90 and can bend singly charged ions of mass 240 at the energy of 40 keV.

(3) Accelerator. The HI-13 tandem accelerator is a High Voltage Engineering Corporation (HVEC) product with four acceleration tubes on both sides of the terminal. Since the accelerator is a heavy ion accelerator, many features are specially designed for good transmission of heavy ions. It is also very suitable for AMS. At present, the maximal terminal voltage can reach 14 MV.

(4) Analyzing magnet. A 90 degree analyzing magnet with a radius of curvature of 127 cm was installed, which has a mass energy product (ME/Z^2) of 200.

(5) Electrostatic deflector. A high-resolution

electrostatic deflector with a radius of curvature of 360 cm and downward deflecting angle of 15 degree was placed on the AMS beam line to reduce the isotopic backgrounds and other unwanted beams.

(6) Detector. At the end of the AMS beam line a detector was installed for final ion identification, which located 2 meters away from the electrostatic deflector. Some detectors, such as $\Delta E-E$ ionization chamber, time-of-flight detector, bragg detector etc., can be used to detect and identify different particles.

2.2 Experimental procedure

The negative ions extracted from cesium sputter source are focused by a trim einzel lens and a double focusing 90 degree deflecting magnet. The first aperture with fixed diameter of 9 mm is located just at the entrance of the pre-acceleration tube. After momentum analysis, the negative ion

beams of interest are then passed through the pre-acceleration tube and accelerated up to the energy of about 120 keV. Then the ions are accelerated with a terminal voltage of about 8 MV. Both carbon foil and gas strippers are available in the head of accelerator. The molecular backgrounds are eliminated due to break-up of molecular ions. After passing through the tank, the positive charged particles with different energy are analyzed by the 90 degree analyzing magnet, having a mass energy product (ME/Z^2) of 200, to eliminate the isotopic background. The high-resolution electrostatic deflector placed on the branch beam line is used further reduce the isotopic background and other unwanted beams. Some kind of detector is installed for final ion identification at the end of the branch beam line.

3 Application Examples

3.1 Biomedicine

Calcium is an important element in human body. It is known that calcium deficiency in human body is related to many diseases such as osteoporosis, arteriosclerosis and so on. ^{41}Ca is a cosmogenic nuclide, which is very rare in nature (the abundance of ^{41}Ca in natural Ca is only 10^{-15}) and long lived ($t_{1/2} \approx 1.0 \times 10^5$ a) with pure electron capture decay emitting Auger electrons and 3.3 keV X-rays. The advantage of the ^{41}Ca tracing with AMS method is that, due to the very low natural levels of ^{41}Ca in biological systems and the long half-life of ^{41}Ca , sensitivities can be increased to meet the requirement of the lowest possible radiological hazard. Therefore, the man-made ^{41}Ca is an ideal tracer in the studies of Ca metabolism, in combination with AMS measurement. It was reported that the radioactive dose received in ^{41}Ca tracing experiment was much less than that from natural radiation^[1]. Research in recent years shows that carcinogenesis is related with free Ca^{2+} in the cells. The level of free Ca^{2+} as a messenger is rather stable in normal

cells. The level of free Ca^{2+} of cytoplasm will increase in the cells exposed to carcinogenic substances such as chrysotile or cigarette smoke solution^[2]. In order to investigate the origin of increased Ca^{2+} , the research on the changes of the free Ca^{2+} in cytoplasm using ^{41}Ca as a tracer is being carried out in CIAE.

The initial research results from the cell stimulation by chrysotile indicated that the origin of the increased free Ca^{2+} in cytoplasm is both extracellular and intracellular^[3].

3.2 Nuclear physics and astrophysics

3.2.1 Half-life of ^{79}Se

There are 1–2 orders of magnitude's discrepancies among the values of ^{79}Se half-life in different literature. The main reason of the large difference is mainly due to the interference of ^{79}Br in AMS measurements. By developing a new detection technique, projectile X-ray Accelerator Mass Spectrometry (PX-AMS), the half-life of ^{79}Se was re-measured in CIAE. ^{79}Se and ^{79}Br were separated directly from their X-ray detection. The new value of the half-life of ^{79}Se is $(1.24 \pm 0.19) \times 10^5$ a^[4].

3.2.2 Source of interstellar ^{26}Al

The source of interstellar ^{26}Al is one of the puzzles in the field of astrophysics. Research shows that, based on today's knowledge, only half of the interstellar ^{26}Al could be produced^[5]. There must be multiple ways to produce ^{26}Al in stars. It was proposed by Peng Qiuhe^[6] that it might be possible to produce ^{26}Al from heavy ions reactions such as C, N and O. The measurements of the cross section for reactions $^{14}\text{N}(^{16}\text{O}, \alpha)^{26}\text{Al}$ and $^{14}\text{N}(^{14}\text{N}, \text{D})^{26}\text{Al}$ have been carried out with projectile energies in the range of 7–20 MeV.

3.2.3 Neutrino measurement

A next generation of nuclear and particle physics experiments will require increasingly pure materials. The signals of extremely rare processes such as neutrino interaction and oscillation, double beta decay, and dark matter interaction are often

drowned out by common terrestrial backgrounds. Radionuclides such as uranium, thorium, and potassium-40 have long half-lives and remain abundant in the earth's crust. The decay products of these nuclei and their daughters can often have the same low-energy signals of the rare processes studied. To enable experiments probing rare phenomena, a key point is to select materials with very high radionuclide purity. A new method based on AMS has been developed for measurement of these trace elements in neutrino detector material. At present, the nuclide ^{40}K in CsI and Liquid Scintillators (LS) has been measured with AMS, and the initial experiment results showed a detection limit of $< 10^{-15} \text{ g } ^{40}\text{K/g}$ may be achieved^[7].

3.3 Hydrological study

For the study of the groundwater age in the deep sediments of Quaternary in Hebei plain, the $N(^{36}\text{Cl}) / N(\text{Cl})$ ratio of groundwater samples were determined by CIAE-AMS. The ^{36}Cl ages were compared with hydrodynamic ages. The age of the groundwater in the third and fourth aquifers of Quaternary sediments in the Baoding district of Hebei plain was found to be relatively young. The third aquifer in the east of Baoding district and the west of Cangzhou district less than 50 thousand

years and that of the fourth aquifer, perhaps more than 100 thousand years. But the groundwater age of the third aquifer in Cangzhou city and Qingxian county was 80—90 thousand years and that in Dongguang county was 260 thousand years. The fourth aquifer in Cangzhou city was 330 thousand years and that in Dongguang county was 770 thousand years^[8].

3.4 Environment monitor

A study on the anthropogenic ^{129}I in the environments around the high radioactivity sites has been carried out in CIAE. The samples were collected from the surface waters and ground waters. ^{129}I concentrations in the samples were measured by using the AMS system. The ^{129}I concentrations in the measured samples around the high radioactivity sites range from 10^8 atoms/L to 10^9 atoms/L, several orders of magnitude higher than its concentration in the original rain water^[9]. So, ^{129}I could be used as an environment monitor around the nuclear sites.

4 Summary and Outlook

Some long-lived radioisotopes successfully measured by CIAE-AMS system are listed in Table 1.

Table 1 Outline of methods and performances of CIAE-AMS

Measured nuclides	Detection method	Detection limit
^{10}Be	Stop absorption and multi-anode ionization chamber	5×10^{-15}
^{28}Al	Multi-anode ionization chamber	1×10^{-14}
^{36}Cl	Multi-anode ionization chamber	3×10^{-15}
^{41}Ca	Multi-anode ionization chamber	1×10^{-14}
^{79}Se	Projectile X-ray	2×10^{-9}
^{129}I	Time-of-flight	1×10^{-13}

At present, the measurement methods of some heavy nuclides such as ^{99}Tc , ^{151}Sm , ^{93}Zr , ^{90}Sr , ^{182}Hf , etc are being studied on the CIAE-AMS system. In order to increase the sensitivity, the next

main objective is to improve the experimental facility which is significant for nuclear waste management and nuclear safe guard.

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中国原子能科学研究院的长寿命核素 加速器质谱测量及其应用*

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摘要: 加速器质谱是近年来蓬勃发展的一种十分活跃的核分析技术, 其应用涉及环境科学、地质学、核物理及天体物理、生物医学等多个领域。中国原子能科学研究院的加速器质谱装置自从 1989 年建立至今, 已经对³⁶Cl, ¹⁰Be, ¹²⁹I, ²⁶Al, ⁷⁹Se 和⁴¹Ca 等多种核素进行了成功测量。介绍了中国原子能科学研究院的加速器质谱计系统、核素的测量原理及几种相关核素的成功应用。

关键词: 长寿命核素; 加速器质谱; 应用

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