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Ultra-sensitive Analysis of Radio-isotopes by Accelerator Mass Spectrometry

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1 Introduction

Accelerator Mass Spectrometry(AMS) [1] is a technique developed in the last decade with an ultra-sensitivity to determine radio-impurity of isotopes to the level of 10^{-15}. This is a level almost impossible to reach by any other existing analytical method. The basic principle of AMS is to accelerate ions to energy on the order of 10 or 100 MeV to get rid of the interference from the unwanted molecules, isobars, or isotopes with mass nearby the target isotope. Very important applications is now quite mature in the field of archeology, geo-science, environment, material science, and life science, etc.

The list of long-lived radio-isotopes routinely measured by the AMS technique includes: ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca and ^{129}I [1, 2]. We are interested to extend this list to new isotopes, and to improve the measurement sensitivities beyond other techniques. In particular, the interesting isotopes to explore are ^{40}K, ^{87}Rb, ^{238}U, and ^{232}Th in liquid scintillator (LS), crystal scintillator (CS), as well as other common detector-related materials.

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A typical AMS facility includes the ion injection system, the accelerator, and the analytical beam line. The ion injection system is responsible for the ion formation of the target material and executes the initial rejection of the unwanted ions to the order of 10^3 from the target one. A reasonable current of the target ion is necessary to be injected into the accelerator at this stage in order to guarantee enough event count rate in the analytical beam line after the acceleration stage. The accelerator will raise the energy of the injected ions to the order of 10 or 100 MeV. At this high energy, the subsequent analytical stages in the analytic beam line can reject the interference of any ions with nearby mass. A typical analyzing magnet after the high energy end of the accelerator can reject the unwanted ions to the order of 10^{-6}. An addition rejection power of 10^{-3} can be achieved by an electrostatic analyzer. The final detector system, such as Time of Flight(TOF) or semiconductor detector, provides the other rejection power of 10^{-2}. So the nominal sensitivity of AMS is on the order of $10^{-14} \sim 10^{-15}$.

2 Application in Neutrino and Astro-Particle Physics Experiments

The biggest experimental challenge facing neutrino and Dark Matter experiments is that on background. The background mostly originates from radioactivity in the environment, in the detector components and from cosmic-ray induced unstable nuclei. In particular, the intrinsic purity of the active detector component is of crucial importance. Big research programs have been set up world-wide to attack these problems on two fronts : (a) making purer materials, and (b) devising methods to measure the major radio-isotopes with better sensitivities.

Some advances have been made in the measurement techniques. However, there are still much room of improvement in the areas of having: (1) shorter measurement time, (2) less sample mass, (3) generic applications to many samples, and (4) probing radio-isotopes which do not emit γ-rays. For instance, the best sensitivity in ^{238}U and ^{232}Th (range of 10^{-15} g /g) was achieved with a one-ton liquid scintillator facility at Gran Sasso Underground Laboratory [3]. It takes large target mass and long measuring time, and the results are only valid for *that* particular LS sample, and assuming secular equilibrium of the decay series. There is much need to devise other methods which extend the capabilities. A simple and effective method to determine the impurity concentration level will therefore have great impact in the field.

There are efforts with high-purity germanium detectors [4], neutron activation analysis, Inductively Coupled Plasma Mass Spectrometry, and direct α-decay counting towards these goals. Every technique has its advantages and drawbacks. For instance, the most commonly-used HPGe are sensitive to radio-isotopes with strong high energy γ-emission, like the ^{238}U and ^{232}Th series and ^{137}Cs, but are not applicable to lone beta-emitters like ^{129}I and ^{87}Rb. In potassium where ^{40}K (with 1.46 MeV γ's)/^{39}K is only 10^{-5}, there are potentials (and strong motivations) to improve on the sensitivities with the different techniques sensitive directly to the ^{39}K.

Accelerator Mass Spectrometry is the most sensitive analytical method known for trace isotopes analysis. Its adaptations to the measurements of radio-isotopes are not well-explored, but we believe there are room to make advances with this technique.

3 Status and Plans

As a pilot test, the TEXONO Collaboration have carried out an AMS run in determining the concentration level of ^{129}I contained in the CsI powder (raw materials for making scintillating crystal CsI(Tl)) in November 2000 with the HI-13 Tandem Accelerator Facility at the China Institute of Atomic Energy (CIAE). The schematics of the facility is depicted in Figure 1. The result was successful. From the test, we have determined that the concentration level of ^{129}I in the CsI(Tl) scintillating crystal for the TEXONO collaboration is on the order of 10^{-13}. This level is difficult to reach technical-wise by other method. What is presented in Figure 2 is the experimental result of the pilot run detected by the TOF detector of the AMS system in CIAE. Plot 1 in the figure shows the result for a standard sample prepared with known the concentration ratio, ^{129}I/^{127}I$\sim 10^{-13}$. Plot 2 is the result for the chemical carrier, called the blank, to extract the ^{129}I from a seaweed sample with a purity level of 10^{-13} also. Plot 3 shows the result of a seaweed sample with 10^{-11} concentration level of ^{129}I. Plot 4 then gives the results for CsI at the level of 10^{-13}.

The successful measurement of this ^{129}I/^{127}I limit demonstrated the capability of the group to pursue further in this program. It would be our goal to develop methods of sample preparation for the determination of the level of ^{40}K, ^{87}Rb, ^{232}Th, and ^{238}U in the LS or scintillating crystal and later on extending to generic samples. None of the current research by the method of AMS is specifically focused on this potential line of applications. We would like to propose a research program along this direction.

The analysis of ^{129}I is a routine application by the method of AMS in that it is the

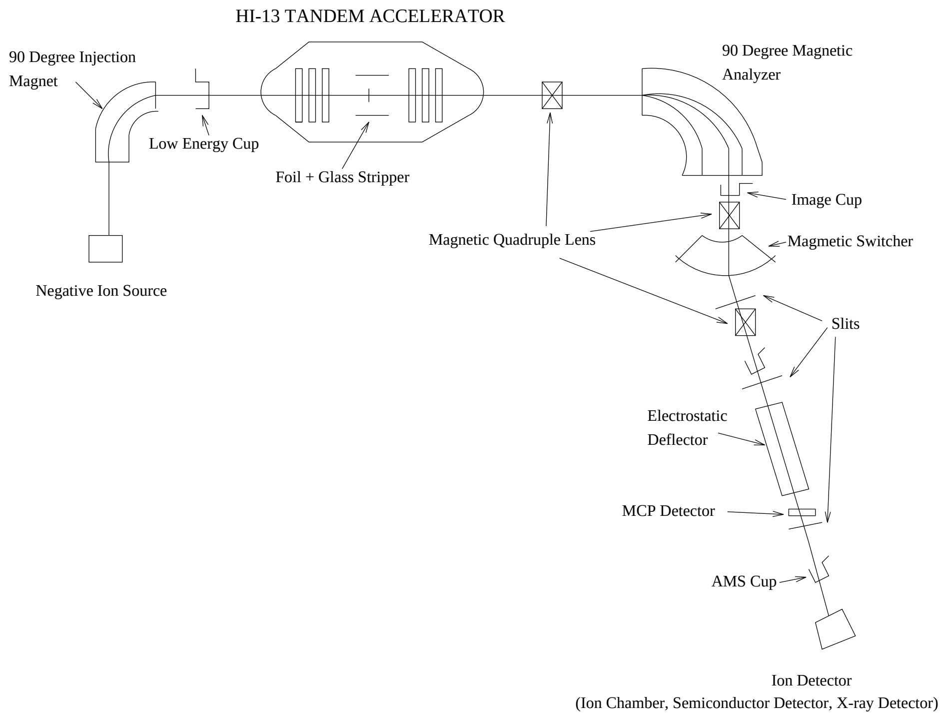

Figure 1: Schematic layout of the 13 MV Tandem Accelerator Facility at CIAE for AMS.

key isotope in the dating of the order of million years in geo-science because of its long half life of 15.7 million years. The method of sample preparation for the ion injection is therefore a common practice for ^{129}I. This is not the case, however, in the measurement for some of the isotopes. The research program is therefore to explore the strategies in the measurement of ^{40}K, ^{87}Rb, ^{238}U, and ^{232}Th with AMS.

- **^{40}K :** The current technique in measuring ^{40}K is quite mature from the viewpoint of preventing from the mass interference of isotopes or isobars. The mass resolution is good enough to resolve ^{39}K, and ^{41}K from ^{40}K, also the isobaric interference from ^{40}Ca can be resolved by using a gas ionization chamber to reject the isobar. It is therefore possible to reach the sensitivity level of 10^{-15}. Another R&D effort has to be emphasized in obtaining a reasonable current out of LS with liquid sample.
  Another indirect method is to measure the concentration of the stable isotope, ^{39}K, in the sample. From the abundance ratio, $^{40}\mathrm{K}/^{39}\mathrm{K} \sim 10^{-5}$, one can then obtain the concentration of ^{40}K in the sample. This method has a potential advantage of higher resolution then the direct measurement. One can determine the level of ^{40}K to better than 10^{-15} as long as ^{39}K is measured to better than 10^{-11}. One, however,

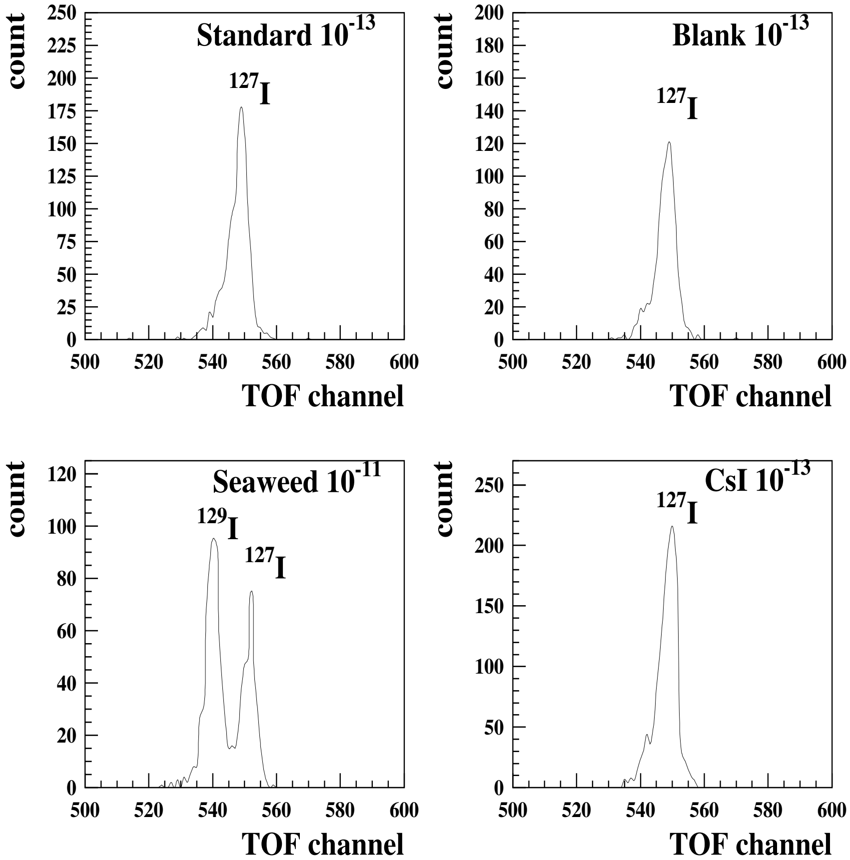

Figure 2: Results of the pilot run in October 2000 at CIAE to measure the ^{129}I/^{127}I ratio in CsI sample.

has to devote a special effort in order to reduce the interference of ^{39}K from the background material since the natural abundance of ^{39}K in the common materials is high. The chemical handling of the sample is critical to prevent the sample from surrounding contamination, given such a high background of ^{39}K abundance.

- ^{87}**Rb :** The stable isotope is ^{85}Rb. There is no serious interference from this isotope. Isobaric interference does exist resulting from ^{87}Sr. One can use X-ray detection technique in the analytical beam line to resolve it. This method, however, is low in efficiency. In similarity to the case of ^{40}K, one can measure the concentration of ^{85}Rb first, and then determine ^{87}Rb from the ratio of natural abundance.

- ^{238}**U and** ^{232}**Th :** The mass interference due to nearby isotopes or isobars is not serious. The sensitivity level of 10^{-10} g/g (best typical HPGe level) from the existing system is not difficult to reach. It requires R&D efforts in the chemical treatment of the sample as well as optimization in the accelerator hardware (better magnets) to reach a higher measuring sensitivity.

The focus of our research efforts in the 2001-2003 time frame would be:

1. Chemical extraction process for the target isotope

2. Prevention of the isotopic contamination from the environment.

3. Ion beam formation of K, Rb, U, Th

4. Injection efficiency calibration.

5. Increase of the ion injection current.

6. The detection efficiency of the heavy isotopes, ^{238}U and ^{232}Th. In order to increase the ion injection current, these heavy isotopes are usually prepared in the form of oxide. This further increases the mass of injection ion. The transportation efficiency will be seriously affected with such heavy ions.

4 Project Organization

This research program will be carried out under the framework of the TEXONO Collaboration which, since 1997, has successfully opened up channels of communication and collaboration among the Taiwan and Mainland China Groups. Channels of movements of research personnel, equipment as well as funding are by now well-established by the Collaboration. This enables efficient and flexible operation and sharing of resources and expertise in the future projects.

The CIAE team (nuclear physics experience) in TEXONO Collaboration is headed by Prof. Zhou Zu-ying. The laboratory has been working with the AMS techniques for ten years with its on-site 13 MV Tandem accelerator [5]. The facility has international-standing in the field and is well-placed to explore new research directions for AMS. Prof. Jiang Shan is the Project Leader of the AMS program. A Ph.D. student (Xin Biao, CIAE) is expected to work on this project with the experts. An independent proposal will soon be submitted by the CIAE team to the National Science Foundation in China to complement this Taiwan NSC Proposal.

The AS team serves as interface of the technique to the actual "market" in low-background experimentation in Dark Matter and Neutrino physics. Wong Tsz-King will co-ordinate the funding allocation as well as the operation of the project within the extended TEXONO Collaboration. Dr. Chen Chin-Ping is the scientific interface person between CIAE and AS.

A new team from NTHU (nuclear science background) joined the Collaboration specially for the AMS project. Their intentions and focus will be to establish contacts with the active players in the field and to gain first-hand experience with the AMS data taking, with the goal of eventually building an AMS facility in Taiwan. The NTHU team will submit an independent proposal for their AMS program.

5 Funding Request

In the 2001-2002 NSC proposal, we submit this AMS-project as a sub-project of the TEXONO research program. That is, the allocated funding will be under the responsibility of TEXONO PI H.T.K Wong.

We request funding for (1) materials cost to explore the techniques of chemical extraction, target preparation, blank control sampling, and calibration (600k NTD), (2) beam time (neutron and heavy ions) operation cost sharing at the Tandem at CIAE (400k NTD), and (3) sharing of equipment (magnets) upgrade to do AMS for heavy isotopes (300k NTD), and (4) travels to Beijing to participate in data taking (5 person-week).

6 Summary

Based on (a) the infra-structures and connections built up by the TEXONO Collaboration in the past few years, and (b) the existing expertise and reputation of the CIAE team in this subject, we propose to explore the use the techniques of Accelerator Mass Spectrometry to enhance and entend the measurements of trace impurities of radio-isotopes. A pilot "demonstration experiment" on the limits of $^{129}I/^{127}I$ ratio was successfully performed.

There are world-wide efforts on this subject with other methods. Success in this project will be both a significant contribution to the advances of the AMS techniques, and also serve as an important standing-point for the Collaboration to pursue the studies of low-energy low-background experiments.

References

[1] "Accelerator Mass Spectrometry", C. Tuniz et al., CRC Press (1998).

[2] "Accelerator Mass Spectrometry for Measurement of Long-Lived Radio-isotopes", D. Elmore and F.M. Phillips, Science **346**, 543 (1987).

[3] G. Alimonti et al., Nucl. Instrum. Methods **A 406**, 411 (1998).

[4] G. Heusser, Nucl. Instrum. Methods **B 58**, 79 (1991);
P. Jagam and J.J. Simpson, Nucl. Instrum. Methods **A 324**, 389 (1993).

[5] "Accelerator Mass Spectrometry with the Beijing HI-13 tandem", S. Jiang et al., Nucl. Instrum. Methods **B 52**, 285 (1990);
"Accelerator Mass Spectrometry at the China Institute of Atomic Energy", S. Jiang et al., Nucl. Instrum. Methods **B 92**, 61 (1994).