AS-TEXONO/00-03
May 10, 2000

Research on the Measurement of Radio-isotopes with the Accelerator Mass Spectrometry(AMS) and the ICP Mass Spectrometry Techniques (Proposal to NSC/AEC 2000)

C.P. Chen, W.P. Lai, H.B. Li, H.T. Wong [α]
Institute of Physics,
Academia Sinica, Taipei, Taiwan.

J.M. Huang [β], S.C. Lu, L.C. Men
Division of Chemical Analysis,
Institute of Nuclear Energy Research, Lungtan, Taiwan.

M. Ho, S. Jiang, Z.Y. Zhou
Nuclear Physics Division,
Institute of Atomic Energy, Beijing, China.

[α] **PI:**

Henry Tsz-king Wong ( ),
Institute of Physics,
Academia Sinica.

[β] **Co-PI:**

Jon-Mau Huang ( ),
Division of Chemical Analysis,
Institute of Nuclear Energy Research.

Abstract

The measurements of trace contaminations of radio-isotopes are very important to Neutrino and Dark Matter experiments. The techniques of Inductively Coupled Plasma Mass Spectrometry and Accelerator Mass Spectrometry offer potential advantages and complementary merits to the other approaches used so far. There are much room of optimizations and experimental innovations to improve on the present sensitivities and efficiencies, as well as to extend the list of measure-able radio-isotopes. We propose a research project to pursue these goals. These efforts will not only play a crucial role in the on-going research program and future directions of the TEXONO Collaboration, and may also be contributions to the advancement of the field of trace radio-purity measurements.

1 Introduction (Proposal Section 15)

The TEXONO [1] Collaboration, made up of more than 40 scientists from major research institutes in Taiwan, Mainland China and the United States and led by the Academia Sinica group, has been involved in a research program relating to the adoption of scintillating crystal detectors in low-energy low-background experiment [1]. The "flagship experiment" is a 500 kg CsI(Tl) detection soon to be installed in Nuclear Power Station II to study low energy neutrino physics [2]. Various R&D projects related to this generic theme have been initiated. Further details and a complete archive of publications, as well as selected photos, transparencies and figures, can be referred to our Web-site at **http://hepmail.phys.sinica.edu.tw/EXONO/index.html/**.

One of the important challenge is to develop techniques to measure contaminations of long-lived radio-isotopes in a variety of samples, and in particular, in the CsI detector. This is essential to this (and indeed *all*) neutrino and astro-particle physics experiments. Until now, radio-purity measurements related to low-background experiments are mainly performed with high-purity germanium detectors [3], and in one case, very elaborate and expensive liquid scintillator facilities underground [4]. The radio-isotopes studied are limited to those from the ^{238}U and ^{232}Th series as well as ^{40}K and ^{14}C.

As the next generation of experiments (on solar neutrinos and Dark Matter, for instance) should achieve lower detector threshold, the existing techniques may be inade-

[1] **T**aiwan **EX**periment **O**n **N**eutrin**O**

quate. In particular, there are several directions where there are strong motivations for improvement:

1. **enhancing the sensitivities further,**
2. **improving on the measurement efficiencies (or reducing measurement time), and**
3. **extending the range of measure-able radio-isotopes.**

These would be important contributions to the future development in the field of low-background experiments - and to the development of trace radio-isotopes measurements as well. Mastering these techniques would put the Collaboration in favorable position in the next-generation efforts, as well as getting established in the world-stage.

Based on these potential significance, we would like to propose a research project to pursue these goals.

2 Experimental Details (Proposal Section 16)

Since 1997, collaborators at the National Tsing Hua University (NTHU) and the Institute of Nuclear Energy Research (INER) have already performed measurements with :

⊙ High-Purity Germanium detectors (HPGe) at NTHU,

⊙ Neutron Activation Analysis (NAA) at INER

⊙ Inductively Coupled Plasma Mass Spectrometry (ICP-MS) [5] at INER,

while plans are made to start an Accelerator Mass Spectrometry (AMS) [6] program at the Institute of Atomic Energy (IAE) in Beijing. These techniques are complementary optimized for different radio-isotopes. 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 gammas)/^{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.

Many samples have been measured with the HPGe systems at NTHU and the NAA technique at INER. Both are standard procedures. The selection of radio-pure materials for the reactor neutrino experiment [2], like the low-activity PMT glass, are based mainly on the HPGe measurements. The typical sensitivity limits of ppb-level are reached for the

^{238}U and ^{232}Th series and the ppm-level for ^{39}K. Samples of CsI(Tl) have been analyzed with the NAA technique. A contamination of 40 ppm of the stable ^{23}Na was observed, while limits were set for the common radio-isotopes.

The measurements of radio-isotopes with the ICP-MS and AMS techniques, on the other hand, are not fully matured and there are much room for R&D efforts. The list of candidate radio-isotopes to be explored include ^{39}K (for deriving the unstable ^{40}K), ^{87}Rb (lone β-emitter), ^{129}I (β-emitter with only low energy γ's), ^{133}Ba (β-emitter with only low energy γ's), and ^{135}Cs (lone β-emitter), as well as ^{238}U and ^{232}Th where improvement beyond the HPGe level is possible.

We have done pilot measurements of CsI samples with the ICP-MS systems at INER [7]. Sensitivities of 1 ppm and 10 ppb have been reached for ^{39}K and ^{85}Rb, respectively. The ^{39}K limits are already comparable to those provided by the HPGe detectors. We plan to extend the measurements to ^{87}Rb and other isotopes, and to improve on the ^{39}K measurements currently complicated by the presence of ^{39}Ar. The current measurements are limited by the "dilution factor" to turn the solid sample into solution form. Potential improvemements are possible by chemistry pre-treatments to reduce the base nuclei (^{133}Cs and ^{127}I) concentration or to enhance the specific radio-isotopes to be probed. R&D efforts to devise these chemistry pre-treatment schemes are necessary to improve the sensitivities. We aim to achieve two orders of magnitude enhancement based on these and other techniques, such that goals are sub-ppb and 10 ppb for ^{87}Rb and ^{39}K, respectively.

In addition, the INER group already achieves ^{238}U and ^{232}Th sensitivities in water to the 10-100 ppt level with ICP-MS. The techniques will be applied to CsI samples. This is a direct measurement of actual ^{238}U and ^{232}Th concentrations, whereas the inferred measurements or limits from HPGe detectors, typically only the ppb range, assume implicitly secular equilibrium.

We have already got approval for 80 hours of beam-time in 2000 to do the first studies of AMS measurements with CsI samples at IAE with the HI-13 Tandem Accelerator facility [8] (approval statement enclosed). Incident energy of 80$-$100 MeV will be used. 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 [6, 9].

The methods for measuring ^{129}I (to the 10^{-13} level for the ^{129}I/^{127}I ratio, or an absolute of 10^6-10^7 atoms) are established and mature. The measurement of ^{129}I with CsI samples will therefore be performed first. It will be followed by pilot studies to measure the other isotopes like ^{39}K and ^{87}Rb and others, which require R&D and optimization efforts to develop techniques such as the chemistry treatment, calibrations and blank control measurements. Sensitivity goals are the ppb levels for these isotopes.

3 Expected Results (Proposal Section 17)

The success of the proposed measurements will be of world-recognized importance to the present and future neutrino experimental program. It will also represent a solid contribution to the advances of the applications of the techniques, particularly AMS. It may have impact to many areas. For instance, future low-background experiments will benefit much if the trace contaminations from a more complete list of long-lived radio-isotopes (a improvement beyond the present capabilities with only ^{40}K and the ^{238}U and ^{232}Th series) can be accurately measured.

The expected results of this proposal would be:

- measurements of ^{39}K and ^{87}Rb in CsI samples with ICP-MS to the 10 ppb and sub-ppb levels, respectively, while measurements on the other candidate radio-isotopes will also start;
- measurements of ^{129}I in CsI samples to the 10^{-13} level with AMS, the team acquiring valuable on-site experience of operating the machine and detectors to perform the measurements;
- completion of pilot R&D studies of measuring ^{39}K and ^{87}Rb and other candidate radio-isotopes with the AMS techniques, identifying and solving experimental problems for future measurements with enhanced sensitivities.

Several publications can be expected from these results, in parallel to the neutrino experiment program.

4 Manpower Distribution and Support (Proposal Section 12)

The ICP-MS as well as other trace analysis facilities at INER to be used for measurements related to this proposal are under the co-ordination of Dr. J.M. Huang ( ) of the

Division of Chemical Analysis, the co-PI of this proposal and contact-person for the INER team.

The AMS measurement at the IAE Tandem accelerator will be led by Dr. S. Jiang ( ) of IAE, who with his group are experts in the AMS technique [8] with an established record. Physicists from the Academia Sinica will participate in the experiment, with the intention of learning the skills such that we can perform the measurements on our own in the future.

The AS group will play the co-ordination role among the collaborating institutes, and to integrate the efforts to the main-stream neutrino and astro-particle physics program of the TEXONO Collaboration.

The facilities for ICP-MS and AMS, as well as the expertise in operation, are provided by the collaborating institutes, INER and IAE, respectively. All the salary cost of research personnel are provided by the respective institutes. In this Proposal, we request funding support for the cost-sharing in AMS-beam time and ICP-MS facility operations, as well as the expenses in materials and other related equipment necessary for the measurements. Travel expenses for AS physicists to IAE for the AMS data taking will be covered by the operation fund from the neutrino program provided by another NSC grant and by the Academia Sinica.

References

[1] "Prospects of Scintillating Crystal Detector in Low-Energy Low-Background Experiments", H.T. Wong et al., hep-ex/9910002, AS-TEXONO/99-02, to be published in Astropart. Phys. (2000).
(Attached in the Selected Publications of PI.)

[2] "A CsI(Tl) Scintillating Crystal Detector for the Studies of Low Energy Neutrino Interactions", H.B. Li et al., hep-ex/0001001, AS-TEXONO/00-01, submitted to Nucl. Instrum. Methods (2000).
(Attached in the Selected Publications of PI.)

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

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

[5] "Principles of Instrumental Analysis", 4th ed., Chapters 11,18, D.A. Skoogm J.J. Leary, Sanuders College Publishing, New York (1992).

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

[7] "The Studies of Determination of Hydrides by Inductively Coupled Plasma Mass Spectrometry", J.M Huang, INER-T1619 (1992);
"Determination of trace amounts of Th-232, U-238 by ICP-MS", S.C. Liu et al., in *Procs. of 1998 Chinese Chemical Society Annual Convention, Gia-Yue, Taiwan* (1998).

[8] "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).

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