# DEVELOPMENT OF ACCELERATOR MASS SPECTROMETRY IN CHINA

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*Abstract*

This paper introduces the developments of AMS and its applications in China. The AMS at PKU has been upgraded recently and a precision better than 0.5% for $^{14}C$ measurement can be reached. Hundreds of samples were dated for the Xia-Shang-Zhou Chronology Project and AMS played an important role in the establishment of chronological framework of those three dynasties. Geological and biomedical studies were also carried out on PKUAMS. AMS at CIAE has measured quite a lot of radionuclides and various detection techniques such as conversed PIXE, gas-filled magnet and gas-filled TOF have been studied. SINR developed an AMS based on a mini-cyclotron and its performance has been improved continuously.

## 1 INTRODUCTION

Accelerator Mass Spectrometry (AMS) is a powerful means to measure long-lived radionuclides with ultra-high sensitivity and has been widely used for the applications of archaeology, geosciences, environmental sciences, biomedical sciences and material sciences. More than 50 AMS facilities distribute around the world now and more than 20 nuclides have been measured by AMS. Most of AMS use tandem accelerator, but there are many special demands to the accelerator techniques, for example the flat-top transmission[1].

AMS has been developed in China since 1980s and there are three AMS facilities at Peking University (PKU), China Institute of Atomic Energy (CIAE) and Shanghai Institute of Nuclear Research (SINR), respectively.

## 2 AMS AT PKU

### *2.1 Facility Upgrade*

Peking University AMS (PKUAMS) facility is based on an EN tandem accelerator and mainly used for $^{14}C$, $^{10}Be$ and $^{26}Al$ measurements[2]. It has been upgraded since 1996 to meet the requirements of the Xia-Shang-Zhou Chronology Project[3]. Fig.1 showed the layout of upgraded set-up of PKUAMS.

Ion source is the most important part in AMS facilities. We choose MC-SNICS ion source with 40 target positions made by NEC. Having carefully checked the design, we found that 25~30 kV is reliable in high voltage endurance but could not meet the beam transmission conditions afterwards. Later, the American design was modified according to our scheme with an additional 20kV acceleration section, which has proved necessary by later experiments and gets good results. In the later testing progress, another shortcoming was found, that the target was not aligned very well and the target wheel was very little eccentrically installed in the ion source. All these problems were resolved by relevant means to get good conditions for high precision measurements.

The injection beam line have been redesigned to be shortened to reduce a few lens and prevent beam emittance extended during transmitting. The dual $30^{o}$ injection magnet was removed from our AMS system to get rid of magnetic fractionation effect. The beam line was realigned from the ion source to the main analysis magnet. Two 1500 l/s turbo pumps were installed at the entrance and exit of EN accelerator to replace 400 l/s pumps. Now the beam transmission efficiency is about 30% including stripping.

We replaced charging belt and voltage-dividing resistors in the EN tandem accelerator, replaced the power supplies for injection, analysis and bending magnets with Danfysik products, as well as the measurement devices for magnetic field. Most of HV power supplies were replaced with Glassman products. Some instruments have installed to stabilize temperature, humidity and AC voltage in the accelerator hall.

A new control system for beam line was constructed with ControlNet, which is a distributing system developed by Group3 Technology Ltd. The system contained 6 local control stations and one PC, which were connected in a fiber-optical loop. The software for the system was developed on the LabVIEW platform and the virtual instruments could be realized. It has fine user interface and is easy to operate. The system is reliable and stable, which control precision can reach 0.05% with a long-term stability of 0.1% per 8 hours [4].

The new data acquiring system consists of two PCs. The front-end PC with NIM units acquires spectra from the particle detector. The master PC is used to read the $^{14}C$ counts from front-end PC and the stable isotope beam current from Keithly 6512, as well as to control the target changing in ion source and the sequential injection of isotopes. The software for this system was also developed on the platform of LabVIEW. The measured results can be written in Excel tables, which is easy to be treated off-line. This system is stable and reliable.

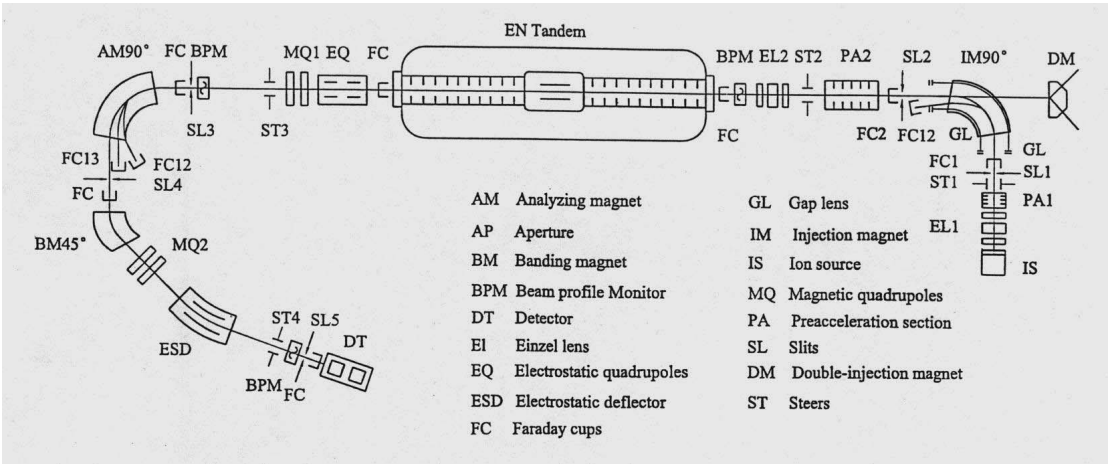


Figure 1: Layout of upgraded set-up of PKUAMS

## *2.2 Radiocarbon Dating*

In order to get high reliable and precision data, the methodology of radiocarbon measurement has been studied deeply.

The stability of ion source, beam transmission, gases in stripper and detector etc. was studied experimentally and improved greatly through these studies[5].

The machine background of the system can reach about 50000 yr for natural graphite by upgrading vacuum of whole system and optimizing system parameters. For reducing the measuring background of radiocarbon samples, different materials and different preparation processes have been studied, especially the influence of the contents of hydrogen element in the prepared sample.

The measuring procedure has been designed carefully. The data of unknown samples were compared with the ones of adjacent standards because the stability of PKUAMS is very good in medium time intervals. Correlation of measured data in each cycle has checked. We also have studied data manipulation method, including abandoning questionable data measured, how to using data of standard samples, and how to calculate internal and outernal errors. At the same time, the method to group samples and to arrange measuring process has been standardized, too. For high precision measurement, one sample was distributed into multiple targets, which were measured individually and then the results were combined if they could pass the t or F test. The off-line data manipulation program (OLDMAP) has been developed on MS Office platform with VBA language, which can manipulate, analyze measured data and finally give $^{14}C$ age and its error.

Several steps have been taken for measuring quality control, such as multi-targets of a sample were measured and their consistency was checked, different standards were measured periodically and the results were compared to their rated values, and the intercomparision between different $^{14}C$ labs have been arranged. It is shown from these data that the results measured by PKUAMS are reliable, usually the deviation from the true value will fall within 2 σ interval.

The measured $^{14}C$ age has to be converted into calendar date using calibration curve and the confident interval will be enlarged during conversion. In order to minimize the effect, calibration of serial $^{14}C$ samples with Bayesian method has been studied[6].

After upgrading the precision of $^{14}C$ measurement of PKUAMS can be better than 0.5%[7].

## *2.3 Applications*

Xia-Shang-Zhou Chronology Project was launched in 1996, aiming to establish the chronological framework of Xia, Shang and Zhou dynasties and radiocarbon dating is one of important means. Hundreds of samples were dated by PKUAMS for the Xia-Shang-Zhou Chronology Project since December 1998, including nine series of samples from the sites Tianma-Qucun, Cemetery of Jin Marquises, Shang city in Yanshi etc[8−10]. AMS played an important role in the establishment of chronological framework of Xia, Shang and Western Zhou.

Lots of AMS biomedical research also carried out at Peking University, such as geonotoxicity study on nicotine and nicotine-derived nitrosamine[11], decay kinetics of nicotine/NNK-DNA adducts[12] and $NaNO_2$'s effects to the toxicity of nicotine[13]. Histone adduction with nicotine[14] and interaction of nicotine and bovine serum albumin[15] have also been studied. Fig.2 gives the decay characteristic of the nicotine-DNA adducts in vivo, which showed two decay stages with different half-life corresponding two different components of the adducts. The experiment was carried out with $^{14}C$-labled nicotine and the extracted DNA was converted to graphite and measured by PKUAMS.

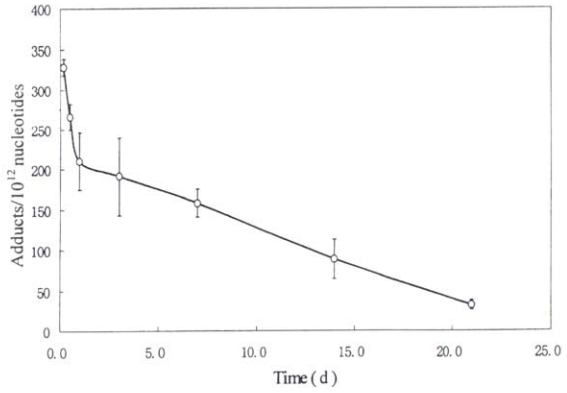


Figure 2: Decay curve of nicotine-DNA adducts in mice liver.

## 3 AMS AT CIAE

AMS at CIAE is based on HI-13 tandem accelerator. Nuclides $^{10}$Be, $^{26}$Al, $^{36}$Cl, $^{64}$Cu, $^{79}$Se, $^{126}$Sn and $^{129}$I have been measured with that facility[16].

### *3.1 Measurement Techniques*

Projectile X-ray AMS (PX-AMS) system has been set up for suppression of isobar interference in $^{79}$Se and $^{64}$Cu measurements[17]. The suppression factor of Br $K_\alpha$ counts is about 250 corresponding to a $^{79}$Se/Se ratio of $3.6\times10^{-9}$, which improved the sensitivity by more than two orders of magnitude comparing with a single Au-Si detector. The overall detection efficiency was $(7.0\pm0.4)\times10^{-4}$ $K_\alpha$ counts per $^{79}$Se ion. The half-life of $^{79}$Se has been determined as $(1.24\pm0.19)\times10^5$ a by PX-AMS, which is quite different with previous reports.

Measurement techniques such as gas-filled Magnet and gas-filled time-of-flight detector[18] have been studied, too. Fig. 3 is a spectrum of $^{58}$Ni and $^{58}$Fe measured by gas-filled magnet, which showed successful separation between the two isobars.

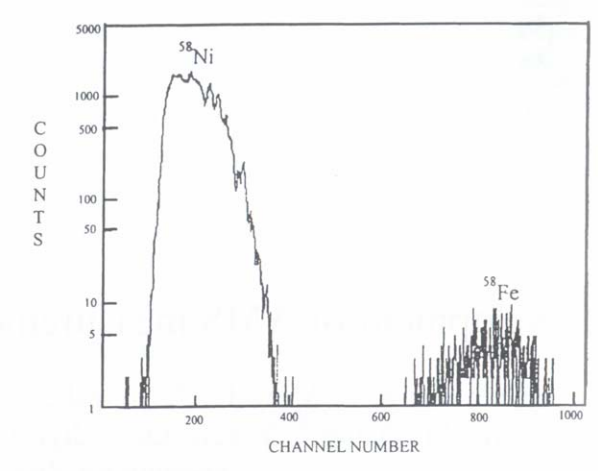


Figure 3: A spectrum of $^{58}$Ni and $^{58}$Fe measured by gas-filled magnet.

### *3.2 Applications*[19]

A project to study the age of deep groundwater in quaternary Hebei plain by AMS $^{36}$Cl method is underway. Samples were taken from the Taihang mountain to seashore in a depth range of 250−550m and $^{36}$Cl/Cl values of $(5.20-1.05)\times10^{-14}$ have been measured. Biomedical applications have been carried out, such as using $^{26}$Al as a tracer to study pneumoconiosis, using $^{41}$Ca as a tracer to investigate the origin of increased free $Ca^{2+}$ in cells when the cells were exposed to carcinogenic substance. Another project is to use $^{79}$Se tracer to study metabolism and bio-dynamics of $^{79}$Sein the body, which is related to many diseases.

## 4 AMS AT SINR

The Shanghai Mini-Cyclotron-based Accelerator Mass Spectrometry (SMCAMS) has been developed at SINR (Fig. 4)[20,21].

### *4.1 Reduction of Fractionation*[22,23]

Various factors, which introduce fractionation, were studied, such as the fluctuation of electrical parameters, the setting of operation parameters, the positioning and cratering of samples, the emittance difference of the injected beams, and the local vacuum near the surface of accelerator parts.

The stray electric field of the deflectors seriously affected the orbit of particles with extra-low energy. After shielding all the deflectors the beam intensity was increased by a factor of two, and the beam stability was improved considerably.

The X-ray produced in mini-cyclotron cannot be discriminated by the MCP detector and introduced a strong background in the total $^{14}$C spectrum. By moving the MCP detector 20 cm above the median plane, the X-ray interference was reduced by a factor of 20.

### *4.2 Refinement of the Methods*[23]

In tandem AMS different carbon isotopes of the same sample are sequentially accelerated, but it is difficult for cyclotron based AMS because most of the power supplies have to be alternately changed by 15%. So other several different sequential acceleration schemes were tested, and finally the adopted one is that only $^{14}C^-$ and $^{13}C^-$ are sequentially accelerated.

### *4.3 Real Sample Measurement*[24]

The $^{14}$C measurement of real samples was carried out in September 1998. The results were satisfactory as compared with those measured by Arizona University and Peking University.

Figure 4: Shanghai Mini-Cyclotron-based Accelerator

## 5 CONCLUSIONS

AMS has developed in China since 1980s and obtained great progress. A lot of nuclides can be measured and innovations have been made in technology. Precision of $^{14}C$ measurements with AMS is better than 0.5%, which can be used for dating in archaeology and geology, as well as for tracing in geoscience and biomedical sciences. It is also fruitful in the applications
Mass Spectrometry (SMCAMS)
with $^{10}Be$, $^{26}Al$, $^{36}Cl$ and $^{129}I$ nuclides. The performance of mini-cyclotron AMS has been improved and first group $^{14}C$ samples have been measured successfully for dating.

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