# Simulation of experimental spectra for medium-heavy nuclides in accelerator mass spectrometry

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**Abstract** Some interferences are often encountered in accelerator mass spectrometry (AMS) measurements, especially for medium-heavy nuclide measurement. It is difficult for online discrimination of the nuclide of interest from the interfering ones. In order to solve this problem, we developed a method to simulate the experimental spectra of medium-heavy nuclides in AMS measurements. The results obtained from this method are in good agreement with experimental values.

**Keywords** Accelerator mass spectrometry, Spectrum analysis, Nuclide measurement, Experimental spectrum
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## 1 Introduction

Accelerator mass spectrometry (AMS) is an ultra-sensitive technique for isotopic analysis. AMS method was developed rapidly for the measurement of long-lived natural radioisotopes in 1970's[1,2]. AMS has now become a powerful and sophisticated analytical tool and been applied widely.

The sensitivity of AMS is mainly limited by the interferences from isobars, isotopes and other backgrounds. These backgrounds cannot be removed completely before they enter into the detector under existing experimental conditions. So it is difficult to discriminate the nuclides of interest from the interferences online in the experimental spectra, especially in the measurement of medium-heavy nuclides with high backgrounds. In the measurements of $^{239}$Pu and $^{182}$Hf performed by Fifield et al [3] and Vockenhuber et al[4] respectively, the high backgrounds caused major difficulties in spectrum analysis. The same problem existed in our measurement of $^{99}$Tc[5]. In order to solve this problem, a method was developed to simulate the experimental spectra of medium-heavy nuclides in AMS measurements. The method can produce a spectrum by considering all kinds of interferences in experiments. The nuclides can be easily discriminated by comparing the calculated spectra with the experimental ones.

## 2 Experimental method

The study based on the AMS facility at the China Institute of Atomic Energy (CIAE) includes three aspects: (1) to analyze the possible interference sources in AMS measurements, (2) to remove and discriminate the interference, and (3) to design the program.

### 2.1 Source of interferences

In the ion source, the negative ion beam is generated from the target material under investigation. Besides the interested nuclide, isobars, isotopes and some molecular ions can also be extracted simultaneously. The interference ions come from: (1) the target material, and(2) the residual gas in the target or the

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ion source, such as carbon, nitrogen, and oxygen. These ions may become interferences in the measurement if they pass through the injector system and the high energy analyzer. Therewith, molecular ions can be broken into fragments when they pass through the stripper (gas or foil). These molecular fragments are the important sources of the continuous momentum spectrum.

The abundant isotopes from the molecular fragments and the continuous momentum from the scattering and charge exchange were suppressed in the high-energy analyzer. For example, if $^{14}$C ion with a charge state of $3^+$ is selected, a potential interference is $^{13}$C from $^{13}$CH since $^{13}$CH has the same mass as $^{14}$C. After stripping, $^{13}$C may have various charge states, some $^{13}C^{4+}$ scattered by the gas of tube may change their charge states and energies. The energies of ions are dependent on the position of scattering, which results in a continuous momentum. Besides the charge changing process, some excitated molecular fragments and ions may decay in the accelerating process, which makes another source of continuous momentum.

## 2.2 Removal and discrimination of interferences

The AMS facility at the CIAE consists of ion source, injector system, tandem accelerator, magnetic analyzer, electrostatic deflector and detectors.

Since isobaric ions have the same mass with the measured nuclide, the removal and discrimination of isobaric ions are dependent on ion source and various detectors. Isotopic backgrounds and molecular interferences are reduced by a sequence of high-resolution momentum, energy or velocity selectors.

After extraction from the ion source, the ions are analyzed with a magnetic spectrometer to choose the ion mass. In tandem accelerator, negative ions are injected and then accelerated to the positive voltage terminal. Then, several electrons are removed in a stripper and the positively charged ions are further accelerated to the ground potential. Molecular ions are dissociated during stripping in the high-voltage terminal. Passing through the accelerator, the ion beam consists of various components with different charge states and different energies. High energy magnetic analyzer selecting the ions with the same $ME/q^2$ suppresses some interferences. An electrostatic deflector is used for selecting the ions with the same $E/q$. At last, ions with the same $ME/q^2$ and $E/q$ as the interested ions enter into the detector.

Usually, $\Delta E$-$E$ method and time of flight (TOF) method are used to identify the ions, which have passed through the electrostatic deflector. The $\Delta E$-$E$ method is based on that the isobaric ions have different energy losses and different residual energies in the gas counter. For example, the method is used to discriminate $^{36}$Cl and $^{36}$S. The method is more effective for the light ions ($Z$<20) with normal tandem accelerator. On the other hand, TOF method is very effective for heavy ions' identification in AMS measurements. It is based on that the isotopes with different masses have different time of flight. For example, $^{129}$I and $^{127}$I can be discriminated by the method.

## 2.3 Program design

After analyzing the sources and the removal process of backgrounds, the simulating program for experimental spectra can be designed. It includes background discrimination, calculation of the values of energy loss or time of flight and then the calculated spectra are obtained.

## 2.4 Discrimination of possible interferences

Most of the unwanted particles are removed by the inject system, high energy analyzer and electrostatic deflector, only the particles with the same $ME/q^2$ and $E/q$ as the interested ions can enter into the detector.

Input parameters of the program include: (1) detailed experimental conditions ($M_1$: extract mass; $M_0$: interested ion mass; $q_0$: charge state; $V_T$: accelerator terminal voltage), (2) parameters of the inject system (mass resolution and $M_1$), (3) magnetic field parameter of high energy magnetic analyzer ($ME/q^2$), and (4) electric field parameter of electrostatic deflector ($E/q$).

The program consists of the following aspects. First, calculating the energy ($E_0$), magnetic field parameter ($C_0$) and electric field parameter ($J_0$) of the interested ions. Second, determining masses of the particles generated in the ion source satisfying $M_2/(M_2-M_0)\geq 80$ or not, which are determined by the mass resolution of the inject system. Third, calculating the magnetic field parameter ($C$) and electric field parameter ($J$) of the particles passing through the inject magnetic analyzer; and then determining the conditions for the particles passing through the high energy magnetic analyzer and electrostatic deflector respectively, namely, $|r_1\text{-}1|\leq 0.01$ and $|r_2\text{-}1|\leq 0.005$ with $r_1=C/C_0$ and $r_2=J/J_0$, which are determined by the resolution of the instruments. Finally, obtaining the mass ($M$), charge state ($q$), and total energy ($E_T$) of ions entering into the detector. The flow chart of the program was demonstrated in Fig.1, where $M_2$ is the mass of ions or molecular ions, which consist of less than three atoms, $X$ is a parameter depending on the mass resolution of the inject magnetic analyzer and $M_1$.

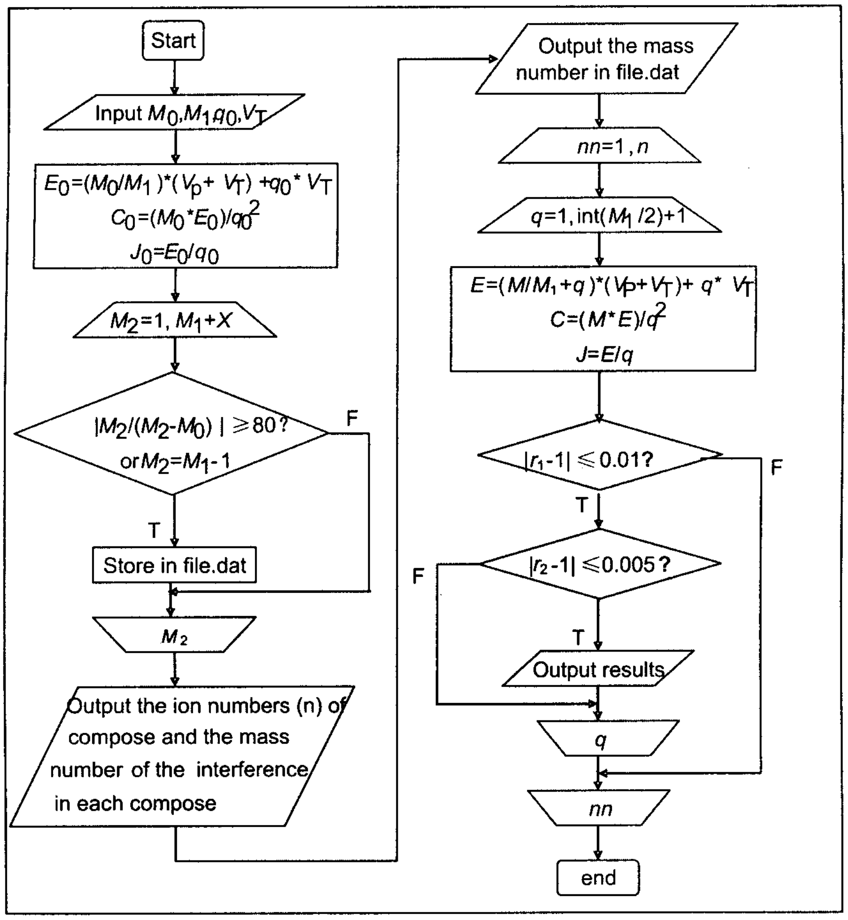

**Fig.1** The flow chart of the program for discriminating the possible backgrounds.

## 3 Results

The theoretical spectra based on $\Delta E$-$E$ detector and TOF detector could be obtained by the program. The procedure of spectrum simulation with the $\Delta E$-$E$ detector was illuminated as follows.

For a given gas pressure and a length of each anode of the multi-anode detector, the energy loss values of the particles entering into the detectors can be calculated. Alpha particles from $^{241}$Am are used for energy calibration of the detectors. Fig.2 gives a flow chart of the calculation.

By inputting the calculated values of energy losses and residual energy the simulated single-dimensional and two-dimensional spectra can be obtained.

The method has been applied in the AMS experiments (e.g. $^{26}$Al, $^{36}$Cl, $^{41}$Ca and $^{99}$Tc). All the tests indicate that the calculated values are in good agreement with the experimental results, and some previously unknown nuclides have been identified. As an example, the simulation results for $^{41}$Ca are given below, in comparison with the experimental values.

In the measurement of $^{41}$Ca, $CaH_3^-$ was extracted from the ion source. The ions were accelerated with a terminal voltage of 7.8 MV. After stripping, $Ca^{8+}$ ions were selected and transmitted to the detector. A multi-anode gas ionization chamber detector was installed for final ion identification. The ionization chamber with a 2 μm Mylar foil window was filled with 13 kPa P10 gas (90% argon plus 10% methane). The calculated values given in Table 1 indicates that the differences of relevant parameters of $^{31}$P, $^{41}$K, $^{36}$S and $^{41}$Ca are very small, so that $^{31}$P, $^{41}$K and $^{36}$S can pass the high energy analyzer and electrostatic deflector and become the possible interferences. Fig.3 (a) and Fig.3 (b) give the calculated and experimental spectra of $^{41}$Ca, respectively. The uncertainty between the experimental spectrum and the calculated one is less than 2% and 3%, respectively. Comparing Fig.3 (a) with Fig.3 (b), the position of $^{41}$Ca and the interferences in the experimental spectra can be easily identified.

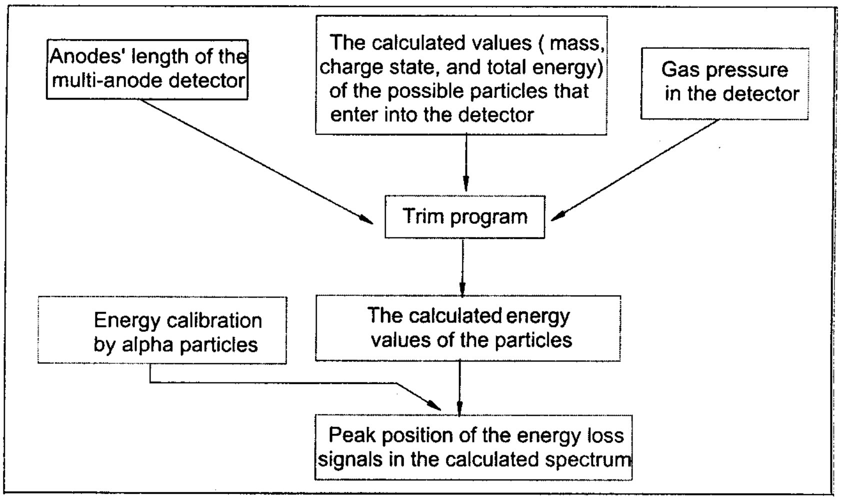

**Fig.2** The flow chart of calculating energy loss and peak position.

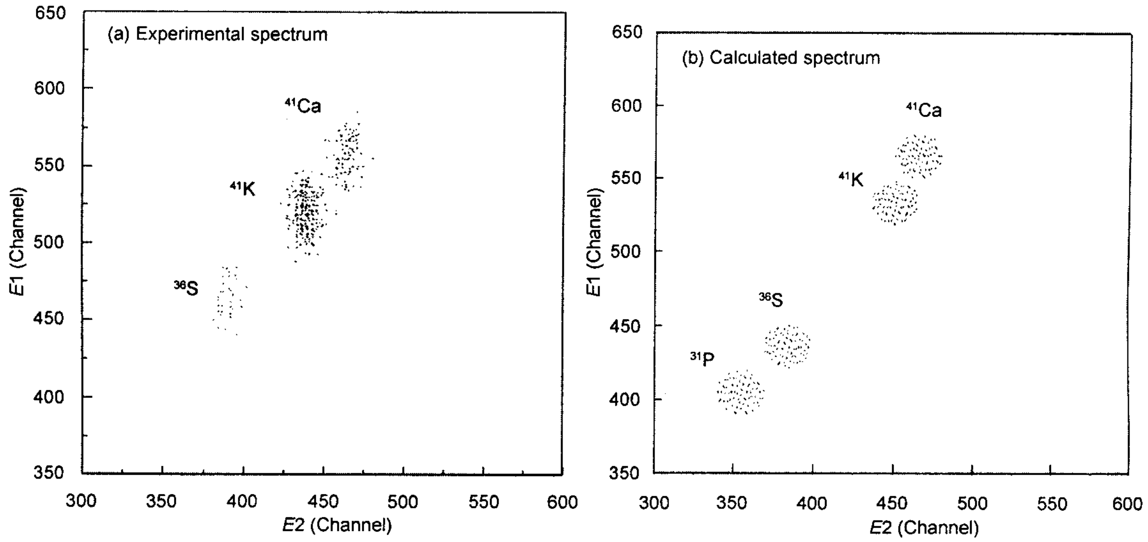

**Fig.3** Comparison between experimental spectrum (a) and calculated one (b).

**Table 1** Calculated values for the ions which maybe enter into the detector

| Particle | *q* | *E* (MeV) | *ME*/*q*$^2$ | *E*/*q* | *m*/*q* |
|---|---|---|---|---|---|
| $^{41}$Ca | 8$^+$ | 69.788 | 44.708 | 8.724 | 5.13 |
| $^{41}$K | 8$^+$ | 69.788 | 44.708 | 8.724 | 5.13 |
| $^{36}$S | 7$^+$ | 60.853 | 44.708 | 8.693 | 5.14 |
| $^{31}$P | 6$^+$ | 51.918 | 44.707 | 8.653 | 5.17 |

There are three interferences ($^{31}$P, $^{36}$S and $^{41}$K) in the calculated spectrum. $^{31}$P does not appear in the experimental spectrum, because differences of the values of *ME*/*q*$^2$ and *E*/*q* between $^{31}$P and $^{41}$Ca are larger than those between $^{31}$P and other two interferences. The other reason is that the concentrations of $^{31}$P in the target material, may be relatively small.

## 4 Conclusions

Judging whether the calculated spectra are in good agreement with the experimental ones should consider two factors. First, the calculated spectra can present all the nuclides existing in the experimental spectra. Second, the relative positions of the nuclides in the calculated spectra are almost the same as the experimental ones.

The calculated values and simulated spectra obtained before the experiment can be used to discriminate the particles in the experimental spectra. The method can shorten the beam time and save the manpower and material resources. The method can also provide the optimized conditions of the experiment, and is suitable for all the particle's measurement at the CIAE HI-13 AMS system. For other AMS systems having different analyzer and different resolution, one should adjust the relevant magnetic and electric field parameters according to the analyzer resolution in order to apply the method more effectively.

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