MASS SPECTROMETRY OF ^{41}Ca WITH RCNP AVF CYCLOTRON

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Introduction

Measurements of abundance ratio ^{41}Ca/Ca are of great advantage for chronology because of the longer half life of ^{41}Ca ($T_{1/2} = 1.1 \times 10^5$ years) than that of ^{14}C which has been used for dating.[1,2] Moreover, since ^{41}Ca isotopes are produced by cosmic rays or neutrons from natural radio-activities near the earth surface, the ratio would be useful for geo- and paleonto-chronology. The expected abundance ratio of ^{41}Ca/Ca is of the order of $10^{-15} - 10^{-14}$. Therefore, the accelerator mass spectrometry (AMS) is at present the only possible method of the measurement.

The first measurement of AMS was performed with a cyclotron by Alvarez and Cornog in 1939.[3] Whereas, the technique has been applied so far mostly with static tandem accelerators. It is mainly due to the fact that the latter accelerator in general accelerates more intense beams than the former, which is very important in spectrometry for samples of very low concentration. However, advantage of using the cyclotron is that it can accelerate positive ions which are sometimes hardly produced with static accelerators, and higher ion energy is obtained, where contaminant of isobars is separated more easily.

We have performed the accelerator mass spectrometry of ^{41}Ca using the AVF cyclotron at RCNP. There are three important points to be achieved; (i) production of calcium ions, (ii) tuning of the cyclotron without monitoring beam intensity, and (iii) identification of ^{41}Ca ions under the intense contaminant ^{41}K ions. In this paper we report briefly on the present status and give perspective of the accelerator mass spectrometry using the cyclotron.

Experimental Procedure

Back-bombard Type PIG Source

An efficient source to produce intense and high charge state of ions is indispensable to the accelerator mass spectrometry for ^{41}Ca. A calcium fluoride (CaF$_2$) single crystal placed on the back of the anode of the PIG source is sputtered with off-resonant xenon ions which are produced in the arc plasma mainly supported by nitrogen gas. The schematical picture of this method is shown in Fig. 1. It was found that very low flow rate of xenon gas was essential to produce higher charge state of calcium ions; for Ca^{7+} the rate had to be kept less than 0.07 cc/min. Typical intensity of the extracted beam was 10 nA for 7+ (147 MeV). Since the RCNP cyclotron is equipped only with this internal ion source, the highest charge state of Ca ions with reasonable beam intensity is 7+ at present. This fact turns to be most serious in lowering the detection limit of the ^{41}Ca/Ca ratio beyond 10^{-11}, which will be described later.
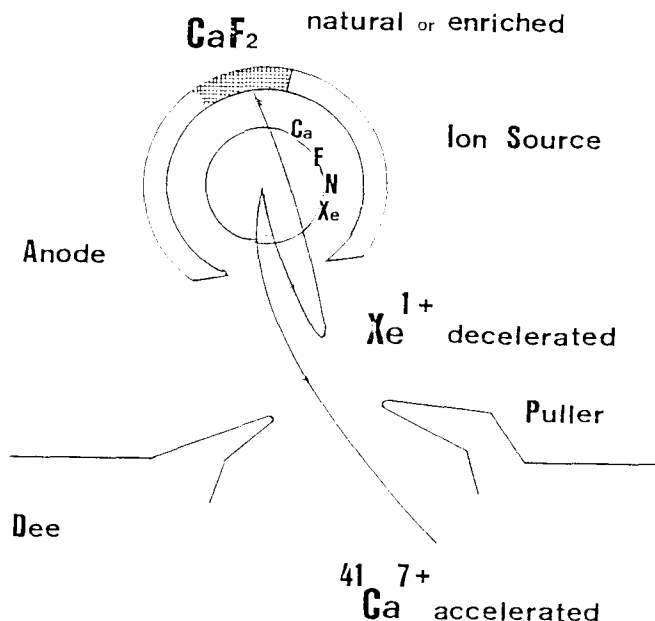

Fig. 1. Schematic picture of back-bombard type PIG source

Acceleration of ^{41}Ca Ions

In the acceleration of ^{41}Ca ions of low intensity, no beam current can be detected by any means. Therefore, at first the cyclotron and the beam transport system have been tuned for ^{40}Ca ions, then, keeping magnetic fields constant, the radio frequency, the acceleration and extraction voltages have been reduced by a factor of the mass ratio 40/41. With this procedure the isochronous orbit, hence, the acceleration efficiency is assured to be the same both for ^{40}Ca and ^{41}Ca.
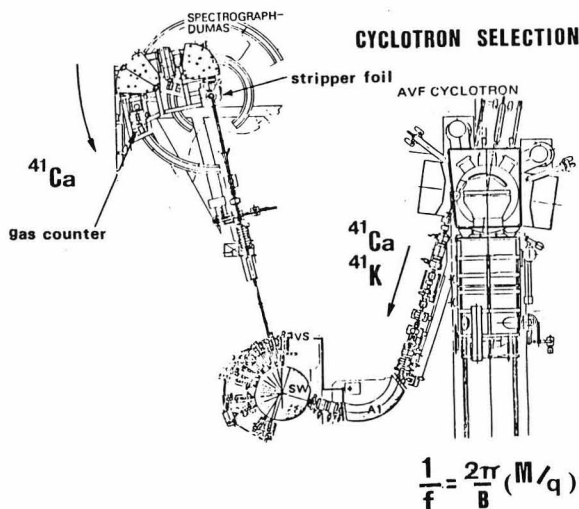
Detection System

Even with the high mass resolving power of the cyclotron, the inherent contaminant of ^{41}K in the crystal (~10 ppm) can not be separated. This contaminant makes a serious disturbance for the ^{41}Ca spectrometry of very low concentration. In order to reduce the ratio ^{41}K/^{41}Ca, charge states of the ions have been selected by using the magnetic spectrograph DUMAS (DUal MAgnetic Spectrograph, QDQDQ).[4] The DUMAS has two focal planes; a dispersive focal plane between two dipole magnets, and an achromatic focal plane downstream the 2nd dipole magnet. A thin stripper foil

of gold was placed at the center of the scattering chamber and the DUMAS was set at 0°. The solid angle of the DUMAS was 5.0 msr, which was large enough to cover the beam passing through the foil. The charge state of the ions was selected with slits at the 1st focal plane. If the charge state of 20+ is selected, all ^{41}K ions are rejected. However, as mentioned before, since the beam energy is not high enough to produce 20+ ions with sufficient intensity, charge states of 19+ or 18+ have been selected, then the ratio ^{41}K/^{41}Ca has been improved by about 1/20 - 1/5. At the 2nd focal plane a detector telescope was placed to identify the ^{41}Ca and ^{41}K ions. The detector consists of a gas ionization chamber (CF$_4$, 50 Torr) as for ΔE detector and a position sensitive Si-E detector. The resolving power of the gas counter was $Z/\Delta Z$(FWHM) $\sim$ 25. Another Si-detector was placed in front of the slit at the 1st focal plane to monitor the acceleration efficiency by detecting ^{41}K ions of lower charge state than that focused on the 2nd focal plane. The schematic illustration of the detection system is shown in Fig. 2.

$$(B\rho)^2 = 2(M/q_{MAX})(E/q_{MAX})$$

MAGNETIC+STRIPPER SELECTION

$$\frac{1}{f} = \frac{2\pi}{B}(M/q)$$

Fig. 2. Schematic illustration of the detection system

Experimental Results

In order to make sure that the present system works well for the mass spectrometry, samples with known concentration of ^{41}Ca/Ca were examined by accelerating Ca^{7+} ions. These samples listed in Table 1. (^{41}Ca/Ca = 1.2×10^{-13} - 1.0×10^{-6}) were prepared by irradiation with thermal neutrons at JAERI. The neutron dose was calibrated by measuring gamma rays in the neutron capture reactions for samples of ^{59}Co and ^{197}Au. The measurements consisted of alternative accelerations of ^{40}Ca and ^{41}Ca. To estimate the beam intensity of ^{40}Ca, elastic scatterings were measured at 10° and the Rutherford cross section was assumed. For ^{41}Ca direct beam counting at 0° was made with the same detection system as for ^{40}Ca. Typically, a few minutes and several tens of minutes of measurements were repeated for ^{40}Ca and ^{41}Ca, respectively. About 10 min. was necessary to switch the accelerating beam from ^{40}Ca to ^{41}Ca and vice versa. In these successive measurements, at least in one cycle, it was assumed that the production and acceleration efficiency of the ions were constant. Further, the same efficiency of charge stripping was assumed for ^{40}Ca and ^{41}Ca ions,

although the beam energy of ^{41}Ca was lower than ^{40}Ca by a factor of 40/41.

Table 1. Samples prepared by thermal neutron irradiation

sample no.	irradiation time	flux density	^{41}Ca/Ca
1	12 hrs.	5.89×10^{13}	1.02×10^{-6}
2	6 min.	2.55×10^{13}	3.66×10^{-9}
13	40 min.	2.58×10^{12}	2.5×10^{-9}
14	5 min.	2.58×10^{12}	3.1×10^{-10}
15	5 min.	1.07×10^{11}	1.3×10^{-11}
16	5 min.	1.09×10^{10}	1.3×10^{-12}
17	5 min.	9.67×10^{8}	1.2×10^{-13}

Due to the contaminant ^{41}K ions, higher charge state is to be selected for samples of lower concentration. However, it was found that the stripping efficiency decreased significantly for higher charge states; the stripping efficiency for the Ca^{20+} ions of $\sim$150 MeV was about 10^{-3} of that of the equilibrium charge state (16.5). Therefore, selection of charge state was made by taking into account both beam intensity and disturbance of the ^{41}K contaminant. For the sample of 1.0×10^{-6} the charge state of 19+ has been selected. However, for the 3.7×10^{-9} sample 18+ has been selected. In this case ^{41}Ca was clearly identified as shown in Fig. 3, although the ratio of ^{41}K/^{41}Ca was about 10^3. For the sample of 1.3×10^{-11} the fully stripped charge state, 20+, has been selected. However, because of the small efficiency only one event has been detected in 64 min. This result seems not reliable since it is likely that the ion source has not been stable in such long duration. In Fig. 4 the concentration of ^{41}Ca/Ca estimated in the present spectrometry is shown. Fairly good agreement has been obtained down to the sample of 3.7×10^{-9}.

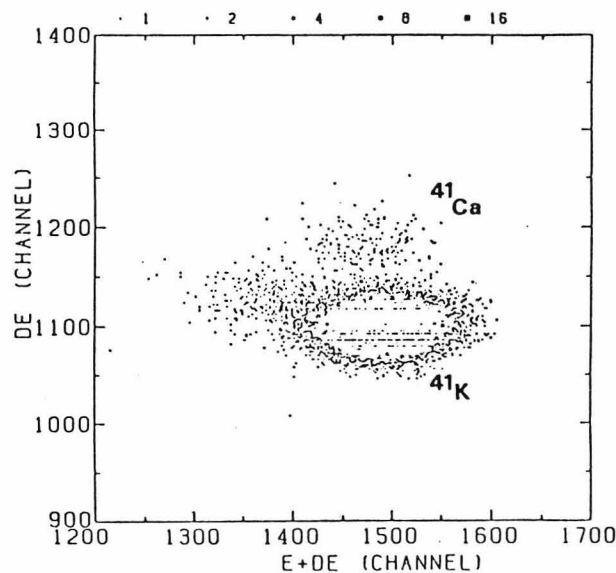Fig. 3. Two dimensional density spectra of dE vs total energy E+dE obtained for a ^{41}Ca-enriched sample (^{41}Ca/Ca = 3.7×10^{-9}).

Perspective of AMS with RCNP Cyclotron

Present limit of the spectrometry is the concentration around 10^{-11}. This is caused by (i) low charge state and small intensity of the ion source and (ii) extremely small stripping efficiency for 20+ ions. Certainly the limit should be extended to the region of terrestrial samples. We are on the way of improving several points; if the higher charge states such as 8+ (192 MeV) are produced sufficiently, stripping efficiency will increase drastically,[5] and some chemical treatments are to be considered to reduce K contaminant in the CaF_2 crystal.

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References

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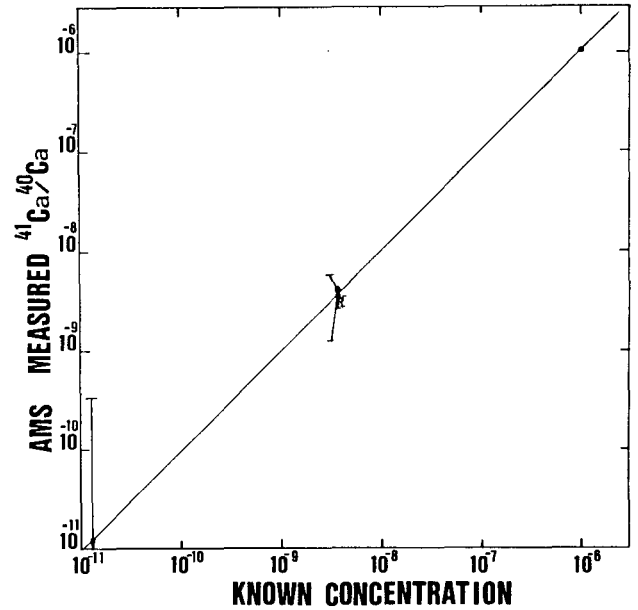4. T. Noro et al., J. Phys. Soc. Jpn. Suppl. 55 (1986) 470.

Fig. 4. The measured ^{41}Ca concentrations versus known concentrations for enriched samples.