[RADIOCARBON, VOL 31, NO. 3, 1989, P 324–326]

AMS OF ^{41}Ca USING THE CaF_3 NEGATIVE ION

PETER W KUBIK and DAVID ELMORE*
Nuclear Structure Research Laboratory,
University of Rochester
Rochester, New York 14627

High-intensity ion sources are now capable of producing negative molecular ion beams of Ca of $>2\mu A$ (Middleton *et al,* 1989). This and the advanced status of ^{41}Ca AMS at several laboratories (Pennsylvania, Argonne National Laboratory, Rochester, Munich, Weizmann Institute) have revived interest in ^{41}Ca ($T_{1/2} = 1.0\times10^5$ yr (Mabuchi *et al,* 1974)) as a possible dating tool (Yamaguchi, 1963; Raisbeck & Yiou, 1979; Kubik *et al,* 1986).

Prerequisites for measuring a radioisotope like ^{41}Ca with AMS are high sensitivity and low background. Using the CaH_3^- molecule from the ion source, the background can be $<1\times10^{-15}$ for ^{41}Ca/Ca (Middleton *et al,* 1989) and the beam currents are high enough (a few μA) (Middleton *et al,* 1989) to measure samples with ratios of ^{41}Ca/Ca between 10^{-14} and 10^{-15}. However, the preparation and storage of high quality CaH_2 samples is not an easy task; sample preparation is best done by a specialist located at the AMS facility. We therefore searched for a different Ca compound with high ion source output and low background in the detector, but with a simpler preparation chemistry.

A mass spectrum of a CaF_2 sample (Urban, Korschinek & Nolte, 1986) showed a surprising similarity with the mass spectrum of a CaH_2 sample (Raisbeck *et al,* 1981). This and the known suppression of potassium from KH_3^- molecules (Raisbeck *et al,* 1981) led us to the investigation of CaF_3^-. We used our Scanning Hiconex ion source (General Ionex Corp, Newburyport, Massachusetts) with an irradiated commercial grade CaF_2 sample. We mixed CaF_2 and Ag in a ratio of 1:4 by weight to insure good heat conductivity for steady beam currents.

The ^{41}K counting rate in the detector from $^{41}CaF_3^-$ injected into the tandem was lower by at least 10^4 compared to injected $^{41}CaF^-$, a reduction factor which is similar to the rejection of ^{41}K when using CaH_2 samples due to the presence of $^{41}KHD^-$. Another critical factor in reducing the background generated by ^{41}K counts in the detector is how well we can differentiate between ^{41}Ca and ^{41}K by measuring several energy loss ΔE and energy E values for nuclear charge Z identification. At a terminal voltage of 10 MV creating 74 MeV mass 41 ions we achieved our usual good separation of ca 5×10^5 with our gas detector using 4 ΔE signals and the energy signal.

We have used a NEC high-intensity ion source (SNICS) with CaH_2 and CaF_2 samples. Typically, 1–2μA of CaH_3^- and 0.7–1μA of CaF_3^- could be produced. With a similar type ion source, Middleton *et al* (1989) could

---

*Present address: Department of Physics, Purdue University, West Lafayette, Indiana 47907 produce 5–10μA of CaH_3^-, so 2μA of CaF_3^- should be possible with that ion source. Disadvantages of using heavy molecules like CaF_3^- are 1) relatively high terminal voltages (10 MV or more) are needed to have enough ion energy for the Z identification in the final detector, and 2) the Coulomb explosion that the molecules undergo in the terminal stripper reduces the transmission through increased emittance. Our measured transmission (including stripper yield for charge state 7) was only 2% using a foil stripper, but we think this can be improved with a gas stripper and other improvements already made to our tandem.

The chemical preparation of samples whether they are bones, rocks or meteorites is straightforward. Samples are dissolved in appropriate acids and the Ca is precipitated as CaO_2O_4 which is converted into CaO by heating at high temperatures. CaO is dissolved in diluted HCl and subsequently Ca is precipitated as CaF_2 by adding HF. After centrifuging, this precipitate is washed several times with distilled water, and then dried in an oven for several hours. CaF_2 is mixed with silver before loading it into a sample holder. This simple procedure will make it possible for an investigator to prepare the samples at his home lab and then send or bring them for the AMS measurements.

Several conclusions can be drawn from this work. KF_3^- shows a very strong suppression relative to the KF^- ion. The beam currents are currently lower than what we can produce from CaH_2, but this might not be a restricting factor for some applications with relatively high ^{41}Ca/Ca ratios. Ratios of 10^{-12} instead of 10^{-15} can be expected in meteorites (Paul *et al,* 1985; Kubik *et al,* 1986; Korschinek *et al,* 1987). Here, ^{41}Ca can be used for exposure history studies and, because of its smaller half-life compared to ^{36}Cl ($T_{1/2}$ = 301,000 yr), can help to determine the terrestrial ages of meteorites in the gap between ^{14}C and ^{36}Cl. Other applications for which the use of CaF_2 might be advantageous are the use of ^{41}Ca as a neutron monitor in uranium ore deposits or as a biological tracer in humans. In this latter application, we could pre-label bones with an oral dose of ^{41}Ca, then monitor the release of ^{41}Ca for many years without the biological damage that would occur from shorter-lived isotopes such as ^{45,47}Ca. It is clear from the recent results of ^{41}Ca in terrestrial surface samples (Middleton *et al,* 1989) with ratios barely above 10^{-15}, that, for these, it is necessary to use CaH_2 because of the higher beam currents available.

ACKNOWLEDGMENTS

We would like to thank N J Conard for his help writing the manuscript and P Sharma for his advice on the sample chemistry. We are grateful to T Miller of the NSRL, and the TAMS group members T K Hemmick, S Jiang and R T D Teng for their help with the accelerator measurements. This work was supported by NSF grant PHY8515908.

REFERENCES

Korschinek, G, Morinaga, H, Nolte, E, Preisenberger, E, Ratzinger, U, Urban, A, Dragovitsch, P and Vogt, S, 1987, Accelerator mass spectrometry with completely stripped ^{41}Ca and ^{53}Mn ions at the Munich tandem accelerator: Nuclear Instruments & Methods, v B29, p 67–71.

Kubik, P W, Elmore, D, Conard, N J, Nishiizumi, K and Arnold, J R, 1986, Determination of cosmogenic ^{41}Ca in a meteorite with tandem accelerator mass spectrometry: Nature, v 319, p 568–570.

Mabuchi, H, Takahashi, H, Nakamura, Y, Notsu, K and Hamaguchi, H, 1974, The half-life of ^{41}Ca: Jour Inorganic Nuclear Chem, v 36, p 1687–1688.

Middleton, R, Fink, D, Klein, J and Sharma, P, 1989, ^{41}Ca concentrations in modern bone and their implications for dating: Radiocarbon, this issue.

Paul, M, Fink, D, Meirav, O, Theis, S and Englert, P, 1985, Determination of ^{41}Ca production for meteorite studies by accelerator mass spectrometry: Meteoritics, v 20, p 726–727.

Raisbeck, G M and Yiou, F, 1979, Possible use of ^{41}Ca for radioactive dating: Nature, v 277, p 42–44.

Raisbeck, G M, Yiou, F, Peghaire, A, Guillot, J and Uzureau, J, 1981, Instability of KH_3^- and potential implications for detection of ^{41}Ca with a tandem electrostatic accelerator: Symposium on accelerator mass spectrometry, Argonne, May 11–13 1981, Proc, p 426–430.

Urban, A, Korschinek, G and Nolte, E, 1986, Ion source study of AMS with ^{41}Ca at the Munich Tandem Accelerator Laboratory: Workshop on techniques in accelerator mass spectrometry, Oxford June 30-July 1 1986, p 108–115.

Yamaguchi, Y, 1963, Possible use of ^{41}Ca in nuclear dating: Progress in theoretical physics, v 29, no. 2, p 567.