Discovery of the Calcium Isotopes

J.L. GROSS and M. THOENNESSEN *

National Superconducting Cyclotron Laboratory and
Department of Physics and Astronomy, Michigan State University,
East Lansing, MI 48824, USA

Twenty four calcium isotopes have so far been observed; the discovery of these isotopes is discussed. For each isotope a brief summary of the first refereed publication, including the production and identification method, is presented.

* Corresponding author.
Email address: thoennessen@nscl.msu.edu (M. Thoennessen).
1. INTRODUCTION

The discovery of the calcium isotopes is discussed as part of the series of the discovery of isotopes which began with the cerium isotopes in 2009 [1]. The purpose of this series is to document and summarize the discovery of the isotopes. Guidelines for assigning credit for discovery are (1) clear identification, either through decay-curves and relationships to other known isotopes, particle or $\gamma$-ray spectra, or unique mass and Z-identification, and (2) publication of the discovery in a refereed journal. The authors and year of the first publication, the laboratory where the isotopes were produced as well as the production and identification methods are discussed. When appropriate, references to conference proceedings, internal reports, and theses are included. When a discovery includes a half-life measurement the measured value is compared to the currently adopted value taken from the NUBASE evaluation [2] which is based on the ENSDF database [3].

2. DISCOVERY OF $^{35-58}$CA

Twenty four calcium isotopes from A = 35 – 58 have been discovered so far; these include 6 stable, 6 proton-rich and 12 neutron-rich isotopes. According to the HFB-14 model [4], $^{63}$Ca should be the last odd-even particle stable neutron-rich nucleus while the even-even particle stable neutron-rich nuclei should continue at least through $^{70}$Ca. At the proton dripline two more isotopes could be observed ($^{33}$Ca and $^{34}$Ca). About 11 isotopes have yet to be discovered corresponding to 30% of all possible calcium isotopes.

Figure A summarizes the year of first discovery for all calcium isotopes identified by the method of discovery. The range of isotopes predicted to exist is indicated on the right side of the figure. The radioactive calcium isotopes were produced using photo-nuclear reactions (PN), neutron capture reactions (NC), light-particle reactions (LP), spallation (SP), and projectile fragmentation of fission (PF).
FIG. A. Calcium isotopes as a function of time when they were discovered. The different production methods are indicated. The solid black squares on the right hand side of the plot are isotopes predicted to be bound by the HFB-14 model.

The stable isotopes were identified using mass spectroscopy (MS). Light particles also include neutrons produced by accelerators. In the following, the discovery of each calcium isotope is discussed in detail.

$^{35}$Ca

$^{35}$Ca was discovered by Äystö et al. in 1985, and published in Observation of the First $T_z = -\frac{5}{2}$ Nuclide, $^{35}$Ca, via Its $\beta$-Delayed Two-Proton Emission [5]. A beam of 135-MeV $^3$He from the Berkeley 88-inch cyclotron bombarded a 2-mg/cm$^2$ natural-calcium target. The $\beta$-delayed two-proton sum spectra were measured and assigned to $^{35}$Ca. “The assignment of the observed groups to $^{35}$Ca is based on excellent agreement with the predicted decay energy for the higher sum peak populating the $^{33}$Cl ground state and with the known energy difference for decays to the ground (G) and the first excited (X) states at 811 keV in $^{33}$Cl. Further, the half-life is consistent with the predictions for $^{35}$Ca and no other new beta-delayed two-proton emitters (e.g., $^{27}$S), if produced, are expected to have these two-
proton sum energies.” The measured half-life of 50(30) ms agrees with the currently adopted value of 25.7(2) ms.

$^{36}$Ca

Tribble et al. first observed $^{36}$Ca in 1976. They reported their findings in *Mass of $^{36}$Ca* [6]. A 131.4-MeV $\alpha$ beam from the Texas A&M University 88-inch cyclotron bombarded a 3-mg/cm$^2$ natural calcium target. The presence of $^{36}$Ca was inferred from the presence of $^8$He detected by an Enge split-pole magnetic spectrograph. “The centroid uncertainty, assuming background contribution, is 30 keV. Combining this with the uncertainties associated with (1) beam energy (10 keV), (2) scattering angle (5 keV), (3) focal plane calibration (15 keV), target thickness (20 keV) along with the $^8$He mass excess of 31.601±0.013 MeV, we find the reaction Q value to be -57.58±0.04 MeV, and the mass of $^{36}$Ca to be -6.44±0.04 MeV.”

$^{37}$Ca

The discovery of $^{37}$Ca was simultaneously reported in 1964 by Hardy and Verrall in *Calcium-37* [7] and Reeder et al. in *New Delayed-Proton Emitters: Ti$^{41}$, Ca$^{37}$, and Ar$^{33}$* [8]. Hardy and Verrall bombarded a calcium target with an 85 MeV proton beam from the McGill synchrocyclotron. The delayed proton spectrum was measured with a surface barrier silicon detector to identify the presence of $^{37}$Ca. “The threshold for production from stable calcium (97% $^{40}$Ca) was found to be 7 MeV higher than that from potassium (93% $^{39}$K), and was approximately 47 MeV. These results are compatible only with the reactions $^{40}$Ca($p,d2n$)$^{37}$Ca and $^{39}$K($p,3n$)$^{37}$Ca, whose calculated laboratory energy thresholds are 44.6 and 38.5 MeV. This establishes the activity as following the decay of $^{37}$Ca.” Reeder et al. used the 60-in. cyclotron at Brookhaven to bombard gaseous $^{36}$Ar with $^3$He at a maximum energy of 31.8 MeV. Proton spectra were measured by two surface barrier detectors. “The excitation function observed for Ca$^{37}$ has a threshold at 20±2 MeV which is consistent with the predicted threshold of 19.4 MeV for the ($^3$He,$^2n$) reaction.” The papers were submitted on the same day and published in the same issue of Physical Review Letters.

$^{38}$Ca

The discovery of $^{38}$Ca was reported in 1966 by Hardy et al. in *Energy Levels of $^{38}$Ca From the Reaction $^{40}$Ca($p,t$)$^{38}$Ca* [9]. A 39.8 MeV beam from the Rutherford Laboratory Proton Linear Accelerator bombarded natural calcium targets. A semiconductor counter telescope was used to detect the emitted particles. The Q value for the $^{40}$Ca($p,t$)$^{38}$Ca reaction was measured and a mass excess was calculated for $^{38}$Ca. “The value obtained for the $^{40}$Ca($p,t$)$^{38}$Ca Q value is -20.459±0.025 MeV.” In 1957 a half-life measurement for $^{38}$Ca of 0.66 s produced in the $^{40}$Ca($\gamma,2n$) reaction was based on the observation of a 3.5 MeV $\gamma$ [10] which could not be confirmed [11]. Another experiment using the same reaction relied on the 1957 measurement and did not identify $^{38}$Ca independently [12].

$^{39}$Ca

$^{39}$Ca was first observed in 1943 by Huber et al.: “Der Kernphotoeffekt mit der Lithium-Gammastrahlung: I. Die leichten Elemente bis zum Calcium” [13]. $^{39}$Ca was populated in a radiative capture reaction with
17 MeV γ-rays. 500 keV protons bombarded lithium to produce the γ-rays from the reaction $^{7}\text{Li}(p,\gamma)$.

Subsequent to the irradiations the decay curves of the emitted β-rays were measured. "Als Resultat von 600 durchgeführten Bestrahlungen erhielten wir die in Fig. 13 aufgezeichnete Zerfallskurve mit einer Halbwertszeit von $T = 1.06 \pm 0.03$ sec." (As a result of 600 irradiations we achieved the decay curve shown in Figure 13 with a halflife of $T = 1.06 \pm 0.03$ sec.). This half-life agrees with the presently accepted value of 859.6(14) ms. A previously reported halflife of 4.5 m [14] could not be confirmed.

$^{40}\text{Ca}$

$^{40}\text{Ca}$ was first observed by Dempster in 1922. He reported his result in *Positive-ray Analysis of Potassium, Calcium and Zinc* [15]. Positive-ray analysis was used to identify $^{40}\text{Ca}$. "With the calcium thus prepared it was found that the component at 44 was still present, and was approximately 1/70 as strong as the main component. We therefore conclude that calcium consists of two isotopes with atomic weights 40 and 44." A year earlier Thomson observed a broad peak around mass 40, however, the resolution was not sufficient to determine which and how many of the isotopes 39, 40 and 41 exist [16].

$^{41}\text{Ca}$

The first identification of $^{41}\text{Ca}$ was described in *A Study of the Protons from Calcium under Deuteron Bombardment* by Davidson in 1939 [17]. CaO targets were bombarded by 3.1 MeV deuterons at the Yale University cyclotron and proton absorption spectra were recorded. A group of protons with a range of 66 cm was attributed to the formation of $^{41}\text{Ca}$. “Since calcium is predominantly $^{40}\text{Ca}$ (96.76 percent), one can almost certainly attribute this group to the reaction $\text{Ca}^{40}+\text{H}_2 \rightarrow \text{Ca}^{41}+\text{H}_1$, giving positive evidence for the actual formation of $\text{Ca}^{41}$."

$^{42,43}\text{Ca}$

Aston observed $^{42,43}\text{Ca}$ for the first time in 1934: *Calcium Isotopes and the Problem of Potassium* [18]. The relative abundances were measured with a mass spectrograph. “Photometry gives the following provisional constitution for calcium: Mass number (Abundance): 40 (97), 42 (0.8), 43 (0.2) 44 (2.3).”

$^{44}\text{Ca}$

$^{44}\text{Ca}$ was first observed by Dempster in 1922. He reported his result in *Positive-ray Analysis of Potassium, Calcium and Zinc* [15]. Positive-ray analysis was used to identify $^{44}\text{Ca}$. “With the calcium thus prepared it was found that the component at 44 was still present, and was approximately 1/70 as strong as the main component. We therefore conclude that calcium consists of two isotopes with atomic weights 40 and 44.”

$^{45}\text{Ca}$

In the 1940 paper *The Radioactive Isotopes of Calcium and Their Suitability as Indicators in Biological Investigations* Walke *et al.* described the discovery of $^{45}\text{Ca}$ [19]. Calcium was bombarded with
8 MeV deuterons at Berkeley and activated samples were placed inside a large expansion chamber. The number of positron tracks on photographs were counted over a 6 month period. A half-life of 180(10) d was observed. “...it is, therefore, probable that this long-lived $\beta$-radioactive calcium isotope is $^{45}\text{Ca}$ produced by the reaction: $\text{Ca}^{44}+\text{H}^2 \rightarrow \text{Ca}^{45}+\text{H}^1$; $\text{Ca}^{45} \rightarrow \text{Sc}^{45}+e^-$.“ The reported half life is in agreement with the currently accepted value of 162.61(9) d.

$^{46}\text{Ca}$

Nier reported the discovery of $^{46}\text{Ca}$ in 1938 in his paper *The Isotopic Constitution of Calcium, Titanium, Sulfur and Argon* [20]. Calcium metal was baked in a small furnace in front of a mass spectrometer and positive ion peaks were observed at 550 °C was used to identify $^{46}\text{Ca}$. “One sees here, in addition to the previously known isotopes 40, 42, 43, and 44, two new peaks, one at mass 48 and one at mass 46.”

$^{47}\text{Ca}$

In 1951 Batzel et al. described the first observation of $^{47}\text{Ca}$ in *The High Energy Spallation Products of Copper* [21]. $^{47}\text{Ca}$ was formed by spallation of copper by 340 MeV protons at the Berkeley 184-inch cyclotron. The existence of $^{47}\text{Ca}$ was determined from the observation of the decay of $^{47}\text{Sc}$. “One was the 150-day $^{45}\text{Ca}$ and the other was a 4.8±0.2-day beta-emitter with an energy of about 1.2 Mev as determined by an aluminum absorption measurement. This activity is probably the 5.8-day calcium activity reported as $^{37}\text{Ca}$ by Matthews and Pool. The growth of a 3.4-day scandium was observed in the decay of the calcium fraction and the scandium daughter was milked from the fraction.” The 4.8(2) d half-life is consistent with the currently accepted value of 4.536(3) d. The mentioned activity by Matthews and Pool was only reported in a conference abstract [22].

$^{48}\text{Ca}$

Nier reported the discovery of $^{48}\text{Ca}$ in 1938 in his paper *The Isotopic Constitution of Calcium, Titanium, Sulfur and Argon* [20]. Calcium metal was baked in a small furnace in front of a mass spectrometer and positive ion peaks were observed at 550 °C was used to identify $^{48}\text{Ca}$. “One sees here, in addition to the previously known isotopes 40, 42, 43, and 44, two new peaks, one at mass 48 and one at mass 46.”

$^{49}\text{Ca}$

$^{49}\text{Ca}$ was first observed by der Mateosian and Goldhaber in 1951, reported in *The Question of Isomerism in Ca* [23]. Enriched calcium was exposed to slow neutrons from the Argonne heavy water reactor and $\beta$-decay curves were recorded following chemical separation. “To our surprise, we were unable to confirm the existence of either of the reported activities when Ca enriched in the isotope of mass 48 (62 percent $^{48}\text{Ca}$) was exposed to slow neutrons from the Argonne heavy water reactor. Instead, we noticed two activities of 8.5 min. and 1 hr. half-life... By chemical separation we could show that the 8.5-min. activity was due to a Ca isotope, $^{49}\text{Ca}$, and the 1-hr. activity due to a Sc isotope, $^{49}\text{Sc}$.” This measured half-life of 8.5(6) m is consistent with the currently accepted value of 8.718(6) m.
The unconfirmed activities mentioned in the quote refer to half-lives of 30 m and 2.5 h reported in 1940 [19].

$^{50}\text{Ca}$

Shida et al. reported the discovery of $^{50}\text{Ca}$ in New Nuclide $^{50}\text{Ca}$ and its Decay Scheme in 1964 [24]. Enriched calcium was bombarded by a 32 MeV triton beam from an electrostatic accelerator in Kawasaki. Gamma-ray spectra were measured at various times following the irradiation. “The weighted average of the half-life is $9\pm2$ sec. Since it was not possible to assign this activity to any known isotopes, it was suspected to be due to $^{50}\text{Ca}$... The results described above seem to be a good basis to attribute the two gamma rays to $^{50}\text{Ca}$. ” The measured half-life is in reasonable agreement with the currently accepted value of 13.9(6) s. A previous attempt to identify $^{50}\text{Ca}$ did not succeed [25].

$^{51}\text{Ca}$

In 1980 Huck et al. described the first observation of $^{51}\text{Ca}$ in the paper $\beta$ Decay of $^{51}\text{Ca}$ [26]. $^{51}\text{K}$ was produced by bombarding uranium with 600 MeV protons at the CERN synchrotron, which decayed to $^{51}\text{Ca}$ through positron emission. Decay curves of $\gamma$-ray spectra were measured. “From the decay of the six strongest lines in the multispectrum, the half-life of $^{51}\text{Ca}$ was found equal to 10.0\(\pm0.8\) s.” This half-life corresponds to the currently accepted value. Only a month later Mayer et al. independently reported the detection of $^{51}\text{Ca}$ by measuring the mass excess [27].

$^{52}\text{Ca}$

$^{52}\text{Ca}$ was discovered by Huck et al. in 1985 and reported in Beta Decay of the New Isotopes $^{52}\text{K}$, $^{52}\text{Ca}$, and $^{52}\text{Sc}$; a Test of the Shell Model far from Stability [28]. A uranium target was fragmented by 600 MeV protons at the CERN synchrotron. Beta-decay curves and $\beta$- and $\gamma$-ray spectra were measured following online mass separation. “A 4.6\(\pm0.3\) s half-life is observed in the decay of other lines (e.g., 675, 961, 1636, and 2070 keV) and is attributed to the activity of the $^{51}\text{Ca}$ parent. This assignment was confirmed by the results of separate multispectrum measurements where the decay of $^{51}\text{K}$ ($T_{1/2}=110$ ms) and the growth of $^{52}\text{Ca}$ ($T_{1/2}=4.6$ s) were simultaneously observed.” The measured half-life corresponds to the currently accepted value.

$^{53}\text{Ca}$

Langevin et al. reported the discovery of $^{53}\text{Ca}$ in 1983 in $^{53}\text{K}$, $^{54}\text{K}$ And $^{53}\text{Ca}$: Three New Neutron Rich Isotopes [29]. Iridium was fragmented by 10 GeV protons from the CERN synchrotron to produce neutron rich potassium isotopes, which then decayed into calcium isotopes. Neutrons were measured in coincidence with $\beta$-rays after the potassium was mass separated. “This work gives evidence for three new K and Ca isotopes and provides further information on half-lives and $P_n$ values.” The measured half-life of 90(15) ms is somewhat smaller than the recent measurement of 230(60) ms, however, the data are consistent with each other within the experimental uncertainties [30].
$^{54-56}$Ca

$^{54}$Ca, $^{55}$Ca, and $^{56}$Ca were first observed by Bernas et al. in 1997, reported in *Discovery and Cross-section Measurement of 58 New Fission Products in Projectile-fission of 750-A MeV $^{238}$U* [31]. Uranium ions were accelerated to 750 A·MeV by the GSI UNILAC/SIS accelerator facility and bombarded a beryllium target. The isotopes produced in the projectile-fission reaction were separated using the fragment separator FRS and the nuclear charge $Z$ for each was determined by the energy loss measurement in an ionization chamber. “The mass identification was carried out by measuring the time of flight (TOF) and the magnetic rigidity $B\rho$ with an accuracy of $10^{-4}$.” 11, 6 and 3 counts of $^{54}$Ca, $^{55}$Ca and $^{56}$Ca were observed, respectively.

$^{57,58}$Ca

$^{57}$Ca and $^{58}$Ca were discovered by Tarasov et al. in 2009 and published in *Evidence for a Change in the Nuclear Mass Surface with the Discovery of the Most Neutron-Rich Nuclei with $17 \leq Z \leq 25$* [32]. Beryllium and tungsten targets were irradiated by a 132 MeV/u $^{76}$Ge ions accelerated by the Coupled Cyclotron Facility at the National Superconducting Cyclotron Laboratory at Michigan State University. $^{57}$Ca and $^{58}$Ca were produced in projectile fragmentation reactions and identified with a two-stage separator consisting of the A1900 fragment separator and the S800 analysis beam line. “The observed fragments include fifteen new isotopes that are the most neutron-rich nuclides of the elements chlorine to manganese ($^{50}$Cl, $^{53}$Ar, $^{55,56}$K, $^{57,58}$Ca, $^{59,60,61}$Sc, $^{62,63}$Ti, $^{65,66}$V, $^{68}$Cr, $^{70}$Mn).”

3. SUMMARY

The discoveries of the known calcium isotopes have been compiled and the methods of their production discussed. The discovery of most of the calcium isotopes was straight forward. Only two isotopes ($^{38}$Ca and $^{39}$Ca) were initially identified incorrectly. $^{37}$Ca is one of the rare cases where two papers reporting the discovery were submitted on the same day.

Acknowledgments

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TABLE I. Discovery of calcium isotopes

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<tr>
<td>Method</td>
<td>Production method used in the discovery:</td>
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<tr>
<td></td>
<td>LP: light-particle reactions (including neutrons)</td>
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<td>MS: mass spectroscopy</td>
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<td>PH: photo-nuclear reactions</td>
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<td>SP: spallation</td>
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<td>PF: projectile fragmentation or fission</td>
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<td>NC: neutron-capture reactions</td>
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<tr>
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TABLE I. Discovery of Calcium Isotopes

See page 11 for Explanation of Tables

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