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Alpha decay of neutron deficient polonium and bismuth isotopes

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ABSTRACT

Neutron deficient isotopes of polonium and bismuth have been produced in the reactions $^{56}\text{Fe} + \text{nat Ce}$ and $^{56}\text{Fe} + ^{141}\text{Pr}$, separated from the primary beam by the gas-filled on-line mass separator, SASSY. The half-lives have been determined by measuring the lifetimes of individual nuclei. The new isotope $^{192}\text{Po}$ ($T_{1/2} = 34\pm3 \text{ ms}, E_{\alpha} = 7.17 \text{ MeV}$) and a short-lived isomeric state in $^{191}\text{Bi}$ ($T_{1/2} = 150\pm15 \text{ ms}, E_{\alpha} = 6.86 \text{ MeV}$) have been identified.

RADIOACTIVITY $^{192, 193, 193m, 194}\text{Po}, ^{191m}\text{Bi}$; measured $T_{1/2}$, $E_{\alpha}$. Gas-filled on-line mass separator.
Heavy ion (HI) induced fusion reactions have become an important means of producing very-heavy as well as very-proton-rich nuclei. The gas-filled on-line mass separator SASSY (Small Angle Separator SYstem, Ref. 1) has been built at the Lawrence Berkeley Laboratory heavy-ion accelerator, the SuperHILAC, to perform the desired fast separation from the primary beam of short-lived reaction products emitted from the target at angles close to 0°. It is possible to study nuclei with half-lives of 0.5 μs or longer with this system.

One difficulty in studying proton-rich nuclei is the competition between neutron evaporation on one hand and fission and charged-particle emission on the other. The use of beams of relatively proton-rich ions such as 56Fe, however, allows for the formation of weakly excited compound nuclei close to the proton-drip line making the effect of the competition from channels other than neutron evaporation less severe.

Neutron deficient isotopes of Po and Bi have been studied previously via fusion reactions induced by ions with A < 40 (Refs. 2, 3, 4). We report here the results of our measurements of the alpha decay properties of 192, 193, 193m, 194Po and 191mBi. We will also discuss the effect on the measured half-lives of accidental correlations between recoil and alpha-particle events. This problem arises since, using SASSY, we measure the life-times of individual recoil nuclei.

In SASSY, the separation of fusion reaction products from the primary beam is achieved by a 1-torr helium-filled magnetic system consisting of a dipole followed by a quadrupole. Downstream from the quadrupole are two gas-filled parallel-plate position-sensitive counters which provide information about the trajectories and energy losses of the recoil
nuclei, and also allow for the determination of their time of flight between the two counters. The recoil nuclei are finally implanted into an array of ten 13 x 22 mm$^2$ Si surface-barrier detectors. The kinetic energy of the recoil nuclei deposited into the Si detectors, together with the counter information, is used in discriminating against unwanted nuclei traversing the separator. The subsequent alpha particle decay of the recoil nuclei imbedded in the detectors is observed as well.

For each alpha event observed in the Si detectors, the identification number of the detector and the arrival time, as well as the time elapsed since the beginning of the previous (or on-going) accelerator beam pulse, are recorded together with the energy information. In addition, for recoil events, the values of eight position and energy-loss parameters from the counters and the time of flight are recorded. Data are stored on magnetic tape using a PDP-15 computer.

The targets were 490 $\mu$g/cm$^2$ nat Ce and 600 $\mu$g/cm$^2$ $^{141}$Pr evaporated on 2050 $\mu$g/cm$^2$ and 1800 $\mu$g/cm$^2$ Havar foils, respectively.

A beam intensity of about $10^{11}$ particles of $^{56}$Fe per second was used for measuring the excitation functions, and the bombarding energy was varied between 242 and 275 MeV.

The alpha-energy calibration was based on known alpha activities produced in the bombardments and on separate calibration measurements using a $^{212}$Pb-$^{212}$Bi-$^{212}$Po source. The activities chosen for internal calibration were $^{192}$Bi (6.06 MeV), $^{191}$Bi (6.32 MeV), $^{195}$Po (6.61 MeV), and $^{194}$Po (6.84 MeV). The energy values are from Ref. 5. When alpha decay takes place inside the detector, the total decay energy is measured except for the pulse height defect which is poorly known for the
recoiling daughter nucleus. The observed energy of an alpha particle from an external source, on the other hand, is affected by the detector window, the thickness of which is uncertain. Because of these factors, we estimate that there is an uncertainty of about 20 keV in the alpha energy values reported in this work. The alpha energy resolution was approximately 50 keV.

We have produced Po isotopes in the reactions $^{\text{nat}}_{\text{Ce}}(56_{\text{Fe}},\text{xn})_{\text{Po}}$ and $^{141}_{\text{Pr}}(56_{\text{Fe}},\text{pxn})_{196-x_{\text{Po}}}$, and Bi isotopes in the reactions $^{\text{nat}}_{\text{Ce}}(56_{\text{Fe}},\text{pxn})_{\text{Bi}}$ and $^{141}_{\text{Pr}}(56_{\text{Fe}},\text{axn or equiv.})_{193-x_{\text{Bi}}}$. The excitation functions for $^{192-196}_{\text{Po}}$ and $^{191-193}_{\text{Bi}}$, including their isomers, and the half-life of $^{192}_{\text{Po}}$ were determined using the $^{56}_{\text{Fe}} + ^{\text{nat}}_{\text{Ce}}$ reaction. Since the counting rate of recoil nuclei in this reaction was too high to allow for the accurate determination of the half-lives of $^{193}_{\text{Po}}$, $^{193m}_{\text{Po}}$, $^{194}_{\text{Po}}$ and $^{191}_{\text{Bi}}$, results from the $^{56}_{\text{Fe}} + ^{141}_{\text{Pr}}$ reaction$^6$ were used for this purpose. As a cross bombardment, we irradiated $^{\text{nat}}_{\text{Ce}}$ with a low-intensity $^{56}_{\text{Fe}}$ beam at 272 MeV in order to confirm the half-life values based on the Pr bombardment.

In Fig. 1 we show an alpha spectrum measured in one of the detectors in the bombardment of $^{\text{nat}}_{\text{Ce}}$ with 254-MeV $^{56}_{\text{Fe}}$ particles. Only alpha particles observed between the accelerator beam bursts are shown and were used in the half-life analysis, since during the beam bursts low-energy, low-mass particles with energy losses too small to produce a signal in the parallel-plate counters, and thus indistinguishable from the alpha particles, were observed in the detectors.

Next we will discuss the method of determining the half-lives of alpha-active isotopes produced in these experiments. In the following,
an "acceptable" recoil nucleus is defined as any recoil product with a kinetic energy and a velocity that falls within limits set to include the majority of the alpha-active fusion products. The procedure is as follows: each alpha particle observed in a given detector is assumed to be the decay signal of the last acceptable recoil nucleus observed in the same detector before the alpha event. In this way, a collection of decay times is obtained for each activity.

This procedure does not necessarily lead to the correct identification of the members of the (recoil nucleus, alpha particle) pair. Accidental correlations become significant whenever the half-life of the activity is not short compared to the average time interval between recoil events in a detector.

There are two possible errors. First, because of the dead time of the analyzer system, it is possible that the correct recoil nucleus was not observed, and thus the wrong recoil nucleus was associated with the alpha particle. These event pairs produce a component in the decay curve with an apparent half-life determined by the counting rate of all acceptable recoil nuclei in the detector in question (the counting rate of alpha particles is low, and can be ignored here). If the number of recoil nuclei within a small time interval follows the Poisson distribution, the apparent decay constant of this artificial component will be equal to \( r \), the average counting rate of acceptable recoil nuclei in the detector.

Another type of error is produced in cases where an alpha decay event observed in a detector was not due to the decay of the last observed recoil nucleus, but belonged to a recoil nucleus observed earlier in the
same detector. In these cases, the observed decay times are shorter than the actual ones. The effect of this phenomenon is given by the equation

\[ \lambda_{\text{obs}} = \lambda + r \]  

(1)

where \( \lambda_{\text{obs}} \) and \( \lambda \) are the observed and real decay constants, respectively.

To illustrate the effect of the accidental correlations, we show in Fig. 2(a) the measured decay curve for the 7.17 MeV alpha activity produced in the reaction \( ^{56}\text{Fe} + \text{nat Ce} \) and assigned to \( ^{192}\text{Po} \). The observed "half-life" of the artificial component is 170±30 ms. The average counting rate \( r \) of acceptable recoil nuclei was 3.94 recoils/s, which corresponds to a half-life of \( \ln 2/r = 176 \) ms in good agreement with the observed value. The correction to the observed half-life of \( ^{192}\text{Po} \) will be discussed below.

In the following, the alpha decay properties of the short-lived \( (T_{1/2} < 1 \text{ s}) \) isotopes \( ^{192-194}\text{Po} \) and \( ^{191}\text{mBi} \) will be discussed. Genetic alpha-alpha correlation measurements have not been possible due to the small alpha decay branches \((< 3.3\%, \text{Ref. 5})\) of the respective daughter nuclei \( ^{188-190}\text{Pb} \) and \( ^{187}\text{Tl} \), and thus the mass assignments are based on excitation function measurements summarized in Fig. 3 and alpha energy and half-life predictions. In Table I, we present the results from our measurements together with previously published data from Refs. 3 and 4.

\( ^{194}\text{Po} \): The excitation function measurements from the reactions \( ^{56}\text{Fe} + \text{nat Ce} \) (Fig. 3) and \( ^{56}\text{Fe} + ^{141}\text{Pr} \) (Ref. 6) indicate that the 6.84 MeV alpha activity belongs to \( ^{194}\text{Po} \). We were not able to confirm the half-life, which is based on results of the \( ^{141}\text{Pr} \) bombardment, using
the natCe cross-bombardment because of interference from the 6.86 MeV 191mBi activity.

193, 193mPo: Results from the excitation function measurements show that the 6.94-7.00 MeV doublet probably belongs to Po with mass number 193. The half-lives of both activities determined from the bombardments of natCe and 141Pr were compatible. We show in Fig. 3 the excitation function for the doublet without resolving it into two components. In those detectors where the components could be resolved, the intensity ratio of the 6.94 MeV and 7.00 MeV alpha lines was found to be independent of the bombarding energy and approximately equal to 0.25. The decay curve for the 7.00 MeV 193mPo from the 56Fe + 141Pr reaction is shown in Fig. 2(b).

192Po: The decay curve for 192Po produced in the bombardment of natCe with 56Fe is shown in Fig. 2(a). The measured apparent half-life, 28.4 ms, was corrected according to Eq. (1) using for \( r \) the experimentally determined value of \( \ln 2/170 \) ms which corresponds to 4.08 recoils/s. The resulting value of 34±3 ms is in good agreement with the value of 32±4 ms determined from the 272-MeV 56Fe + natCe bombardment, where the counting rate of recoils was negligibly low. Further confirmation for the 192Po mass assignment is given by the excitation function calculated for the reaction 140Ce(56Fe,4n)192Po with the neutron evaporation code JORPLE7. The calculated value for the energy of the peak of the excitation curve is within 2 MeV of our experimental value, as can be seen from Fig. 3. The shape of the experimental excitation function strongly suggests that no charged particle emission is
involved in the formation of this evaporation residue. The peak cross section for the reaction $^{140}\text{Ce}(^{56}\text{Fe},4n)^{192}\text{Po}$ is of the order of 10 $\mu$b.

$^{191}\text{Bi}$: The bombarding energies used in our experiments were not high enough to allow for the determination of the excitation functions for the 6.32 MeV, 6.63 MeV, and 6.86 MeV activities, which have all been assigned to Bi with mass number 191 in previous investigations$^3$; however, the threshold energies for all three activities were found to be similar, and the relative yields were within a factor of 2 from those reported in Ref. 3. We therefore conclude that all three activities probably belong to Bi with mass number 191. The half-life of 150±15 ms measured for the 6.86 MeV activity differs significantly from the value of 20±15 s reported in Ref. 3. Our upper limit for the intensity of a component in the 6.86 MeV peak with $T_{1/2} \geq 5$ s is approximately 50% of the intensity of the 6.63 MeV peak. In Ref. 3, the relative intensities of the 6.63 MeV and 6.86 MeV peaks were reported to be 30% and 70%, respectively. In this work, the relative intensities were found to be 25% (6.63 MeV) and 75% (6.86 MeV). We were unable to determine the half-life of the 6.63 MeV activity accurately, but it is on the order of seconds.

The half-lives measured for $^{193,193\text{m},194}\text{Po}$ in this work are consistently shorter than the previous values. There is no previously published value for the half-life of $^{192}\text{Po}$. Our result, 34 ms, is in good agreement with the value of 30 ms calculated from the formula of Taagepera and Nurmi.$^8$ Of the mass formulas published in Ref. 9, the one by Liran and Zeldes reproduces the experimental trend in the Po alpha energies most closely, although the predicted energies are systematically higher.
than the experimental ones. Our value for the $^{192}$Po alpha energy, 7.17±0.02 MeV, is compatible with the systematics of Liran and Zeldes. It is, however, significantly different from the value of 7.12±0.02 MeV of Ref. 4. Our value of 150±15 ms for the half-life of $^{191}$mBi is grossly different from the previously reported$^3$ value of 20±15 s and also from the observed half-life of the 6.63 MeV activity previously$^3$ assigned to $^{191}$mBi. Thus the 6.63 MeV and 6.86 MeV transitions cannot depopulate the same level, contrary to the findings of Ref. 3.

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Footnotes and References

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9. S. Maripuu (ed.), Atomic Data and Nuclear Data Tables 17, Nos. 5-6, 476 (1976).
TABLE I. Results of alpha decay measurements

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Present Work</th>
<th>Ref. 3, Ref. 4</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>$E_{\alpha}$ (MeV)</td>
<td>$T_{1/2}$ (ms)</td>
</tr>
<tr>
<td>-----------</td>
<td>--------------------</td>
<td>----------------------</td>
</tr>
<tr>
<td>$^{194}$Po</td>
<td>6.84$^a$</td>
<td>410±30</td>
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<tr>
<td>$^{193}$Po</td>
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<td>360±50</td>
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<tr>
<td>$^{193}$mPo</td>
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<td>260±20</td>
</tr>
<tr>
<td>$^{192}$Po</td>
<td>7.17±0.02</td>
<td>34±3</td>
</tr>
<tr>
<td>$^{191}$Bi</td>
<td>6.86±0.02</td>
<td>150±15</td>
</tr>
</tbody>
</table>

$^a$ Value used for internal calibration.

$^b$ Ref. 4.

$^c$ Ref. 3.
Figure Captions

Fig. 1. Alpha particle energy spectrum of the nuclei produced in the reaction $^{56}\text{Fe} + ^{\text{nat}}\text{Ce}$ ($E_{\text{lab}} = 254$ MeV). Events observed during the beam pulses have been excluded.

Fig. 2(a). The decay curve of $^{192}\text{Po}$ produced in the reaction $^{56}\text{Fe} + ^{\text{nat}}\text{Ce}$. The artificial long-lived component results from accidental correlations between recoil and alpha particle events.

Fig. 2(b). The decay curve of $^{193m}\text{Bi}$ produced in the reaction $^{56}\text{Fe} + ^{141}\text{Pr}$.

Fig. 3. Relative yields of Po and Bi isotopes produced in the reaction $^{56}\text{Fe} + ^{\text{nat}}\text{Ce}$. The open and solid arrows show the estimated locations of the maxima of the xn curves for the reactions $^{56}\text{Fe} + ^{140}\text{Ce}$ (88.5% abundance) and $^{56}\text{Fe} + ^{142}\text{Ce}$ (11.1%), respectively. The calculated excitation function for the $^{140}\text{Ce}(^{56}\text{Fe},4\alpha)^{192}\text{Po}$ reaction is not to scale, but the experimental curves are directly comparable.
Fig. 1
(a) 7.17 MeV $^{192}\text{Po}$
34±3 ms

(b) 7.00 MeV $^{193m}\text{Po}$
260±20 ms

Fig. 2
Fig. 3
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