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January 19, 1967

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ABSTRACT

Isotopes of francium lighter than mass 214 were studied at the Heavy Ion Linear Accelerator by bombardment of ^{197}Au with ^{16}O and of ^{205}Tl with ^{12}C . Si(Au) surface-barrier detectors were used in on-line measurements to determine α -decay energies and half-lives. Mass number assignments of francium 213 through 203 were based on excitation functions and on genetic relationship with emanation and astatine isotopes. Isomerism was detected in ^{204}Fr . Results are compared with previous work by Griffioen and Macfarlane.

I. INTRODUCTION

In previous work neutron-deficient francium isotopes of mass number 204 through 213 were produced in reactions induced by complex nucleon projectiles accelerated in the Berkeley Heavy Ion Linear Accelerator (HILAC) by Griffioen and Macfarlane who determined α energies, half-lives, and excitation functions for those isotopes. In several cases, daughter activities were separated and identified to obtain supporting evidence for the assignments.

Since that work, the performance of the HILAC and the experimental techniques have been much improved and we have undertaken a study of the light isotopes of emanation,² astatine,³ radium,⁴ and actinium.⁵ As part of this program we obtained additional data on francium isotopes to check and extend the previous work. We also considered it important to look for isomerism in the light francium isotopes because of the systematic occurrence of isomerism in emanation,² astatine,³ and polonium.^{6,7,8}

Excitation functions were determined both for the francium isotopes and for their α -active daughters. These curves made it possible to relate specific francium activities very clearly to daughter isotopes of emanation and astatine. The isotopic assignments of Griffioen and Macfarlane were confirmed. However, our production rates were one or two orders of magnitude higher so that in general we were able to measure half-lives and α -particle energies with more accuracy. Some of our half-life results are significantly different from the previous report. A new isotope, ²⁰³Fr, was detected, and ²⁰⁴Fr was found to have an isomeric state.

II. EXPERIMENTAL

The on-line techniques used in the present study were modifications of those used by Griffioen and Macfarlane.¹ Essentially, the products recoiling from a thin target were thermalized in a helium atmosphere and swept through a small nozzle onto a catcher foil in an adjacent vacuum chamber. The products thus deposited were then quickly positioned in front of a Si(Au) surface-barrier detector. Small amounts of activity were collected on the catcher foil in less than 50 msec while the collection time for the major part was of the order of seconds. In a typical experiment recoil activity was alternatively collected for a fraction of a second and counted for the same period, with the cycle repeated over a period of 10-20 minutes. The apparatus, electronics, and the special energy calibration method are described in a previous article.³ The energy standards used to calibrate the α spectra are given in the heading of Table I.

The reactions used in this study were

where x refers to the number of evaporated neutrons. The targets were 2.5 mg/cm² gold leaf, and separated ²⁰⁵Tl (99% ²⁰⁵Tl, 1.0% ²⁰³Tl, according to the supplier, Oak Ridge National Laboratory) evaporated on 2.3 mg/cm² nickel foil. Beam energy was varied by degradation through stacks of aluminum absorber foils as described earlier.³ The range-energy data of Northcliffe⁹

were used. Maximum beam energy was 166 MeV for oxygen, 126 MeV for carbon. Beam currents were of the order of 0.5 μ A.

It was noted earlier³ that the collection efficiency for astatine, but not for francium, could be increased by an order of magnitude by mixing 1% air with the helium in the stopping chamber. We took advantage of this to obtain definite excitation functions for the astatine isotopes.

In the case of $^{197}\text{Au} + ^{16}\text{O}$, the excitation functions were measured starting at the Coulomb barrier and the beam energy was progressively increased to its maximum. The total integrated beam current was the same for all runs, each of which required about 12 min, with an interval of about three minutes between measurements. With this time schedule, some distortion of the excitation functions of the longer-lived activities was observed because residual activity from one measurement was recorded in the succeeding run. For the $^{205}\text{Tl} + ^{12}\text{C}$ case, we started at maximum beam energy and progressively decreased the energy until the barrier was reached.

In a separate series of experiments, we studied the emanation isotopes produced as electron capture daughters of francium. These products were studied with apparatus described by Valli, Nurmiä, and Hyde,² in which the gaseous emanations diffused in vacuum from a target chamber to a second chamber where they were observed with a Si(Au) surface-barrier detector.

In separate experiments half-lives of individual alpha peaks were determined with the techniques outlined in a previous article.³

III. RESULTS

Alpha spectra of the products obtained when ^{205}Tl was bombarded with ^{12}C ion beams of three energies are shown in Fig. 1. Excitation functions derived from similar spectra taken at 24 bombarding energies are shown in Fig. 2. Alpha spectra of the products obtained when gold was bombarded with ^{16}O beams of three energies are shown in Fig. 3. Similar spectra were obtained at 16 other beam energies and they were used in the construction of excitation functions for francium (Fig. 4) and astatine (Fig. 5). Some of the excitation curves in Figs. 2 and 4 are composites for pairs of francium isotopes whose α -particle energies are too close for clear resolution with the silicon detectors. The evidence on which the mass assignments are made is discussed later. Here we comment only that the francium/excitation curves have the typical appearance of those for compound nucleus reactions.

In the excitation functions for the astatine isotopes presented in Fig. 5, the isotopic labels are based on the agreement of the observed alpha energies and half-lives with published values for astatine isotopes.^{3,10} In our α spectra the astatine peaks are more prominent than in the work published by Griffioen and Macfarlane, owing to the presence of air in the helium stopping gas. A large part of the astatine activity is produced by the α decay of francium parents and, as expected, each astatine isotope has a maximum at the same beam energy as the corresponding francium parent. However, the astatine curves in Fig. 5 have more or less clear second maxima at higher bombarding energies. We may possibly attribute the presence of these second maxima to direct production of astatine. We shall consider the significance of these second maxima in the astatine curves in the discussion section. The other details of the

excitation functions are discussed in the following paragraphs in connection with the individual francium isotopes.

Alpha spectra of emanation isotopes produced in the bombardment of gold with ^{16}O and isolated by the methods of Valli, Nurmi, and Hyde² are shown in Fig. 6 and the corresponding excitation functions are shown in Fig. 7. The intensity maxima of the emanation curves fall at the same ion beam energies as the maxima shown in Fig. 4 for their francium parents. The emanation isotopes are formed by electron capture decay. The similarity in the curves indicates that direct production of emanation is negligible.

A compilation of our francium results is given in Table I. The quoted errors are conservative limits covering the total spread in several individual measurements and are substantially greater than the statistical uncertainty of those measurements.

Francium-213. An α peak at 6.773 MeV (Fig. 1) belongs to ^{213}Fr as established in the earlier report.¹ Our excitation functions for ^{213}Fr and for its electron capture daughter, 19 msec ^{213}Em (Fig. 2), support this. Our measurements provide the following values for several properties: ^{213}Fr half-life, 34.7 ± 0.3 sec; ^{213}Fr electron capture decay branching, $0.57 \pm 0.03\%$; ^{213}Em α -particle energy, 8.090 ± 0.008 MeV.

Francium-212. Alpha groups with energies between 6.26 and 6.41 MeV appear in the spectra (Fig. 1) and have the expected excitation functions for ^{212}Fr (Fig. 2). The α -energy values agree with the reported values for ^{212}Fr .¹⁴ However, our measurements do not reveal the full complexity of the ^{212}Fr α spectrum, the subject of a separate study to be published in another paper.¹⁵

Francium-211 and Francium-210. An α peak at 6.54 MeV is seen in Fig. 1. Its excitation function in Fig. 2 is too broad for a single mass assignment. We attribute it to ^{211}Fr and ^{210}Fr , as did the previous authors.¹ At 80 MeV beam energy, where 6.533 MeV ^{211}Fr should predominate, we obtained a half-life of 3.06 ± 0.06 min. At 107 MeV, where the principal activity is 6.537 MeV ^{210}Fr , we measured a half-life of 3.16 ± 0.06 min which does not agree well with Griffioen and Macfarlane's value of 2.65 ± 0.08 min. Our initial counting rate was a factor of 200 greater. As an additional check, we used the reaction $^{197}\text{Au}(^{16}\text{O}, 3n)^{210}\text{Fr}$ at 80 MeV, at which beam energy ^{211}Fr could hardly be produced. The half-life obtained in this case was 3.20 ± 0.06 min.

Francium-209 and Francium-208. The maximum in the excitation function of the 6.647 MeV α peak in Fig. 4 is somewhat broad for a single mass number. We agree with the previous work which assigned this peak to ^{209}Fr and ^{208}Fr .¹ At an ^{16}O beam energy of 80 MeV, where the 6.647 MeV activity should be nearly pure ^{209}Fr , we observed a single half-life value of 52 ± 2 sec. At several beam energies between 100 MeV and 120 MeV, where ^{208}Fr should contribute nearly all the 6.647 MeV α activity, we obtained a half-life of 59 ± 2 sec, which is much longer than the 37.5 sec figure reported by Griffioen and Macfarlane.¹

New evidence for these assignments comes from the excitation functions of the astatine daughters. In Fig. 5 the peak of the ^{205}At curve occurs at 96 MeV, indicating that its α parent ^{209}Fr has its maximum production near this beam energy. In the case of ^{204}At the maximum in its excitation function is 106 MeV, which argues that its α parent, ^{208}Fr , has maximum yield near this beam energy.

Francium-207 and Francium-206. A broad peak at 6.78 MeV can be seen in the α spectrum in Fig. 3. Using a high resolution detector, Griffioen and Macfarlane were able to resolve this peak partially into components assigned to ^{207}Fr and ^{206}Fr .¹ With careful calibration, we determined α energies of 6.773 ± 0.005 and 6.792 ± 0.005 MeV for ^{207}Fr and ^{206}Fr , respectively. The excitation functions of these partially resolved peaks, shown in Fig. 4, support this assignment, as do the excitation functions of the astatine daughters in Fig. 5. The ^{203}At curve has its first maximum at 116 MeV beam energy. The two peaks at 6.133 and 6.227 MeV belonging to ^{202}At and ^{202m}At both have their first maxima at 125 MeV beam energy. These positions correspond reasonably well with the maxima of their respective α parents in Fig. 4.

At 130 MeV beam energy, where ^{206}Fr should contribute predominately to the 6.78 MeV α group, we obtained a half-life of 15.7 ± 0.3 sec. At 106 MeV, where ^{207}Fr is dominant, we measured 14.7 ± 0.3 sec. Since this is somewhat shorter than the 18.7 sec value reported earlier,¹ we remeasured the half-life with a ^{207}Fr sample prepared by the $^{197}\text{Au}(^{20}\text{Ne}, 6n)^{211}\text{Ac} \xrightarrow{\alpha} ^{207}\text{Fr}$ reaction at 139 MeV beam energy. The $^{197}\text{Au}(^{20}\text{Ne}, 7n)^{210}\text{Ac} \xrightarrow{\alpha} ^{206}\text{Fr}$ reaction is weak at this energy. The value obtained in this case was 14.4 sec.

Additional support for the francium assignments comes from the emanation daughter excitation functions shown in Fig. 7, if allowance is made for the fact that the α groups of ^{207}Em and ^{208}Em and those of ^{206}Em and ^{205}Em are not resolved.

Francium-205. The α peak at 6.917 MeV (Fig. 3) with a half-life of 3.7 second was assigned to ^{205}Fr by Griffioen and Macfarlane on the basis of its excitation function and on the observation of ^{201}At , the α daughter of ^{205}Fr .¹

Our observations agree with this assignment, but our evidence on the behavior of the ^{201}At daughter activity (Fig. 5) was different from that reported by the above authors. The ^{201}At production threshold is the same as for ^{205}Fr , and there is a shoulder at the beam energy corresponding to the ^{205}Fr production maximum, but the yield of ^{201}At continues to rise rapidly as the ^{16}O beam energy is increased. At the higher beam energies, the major fraction of the ^{201}At is probably produced by some direct mechanism.

We have additional evidence in support of the ^{205}Fr assignment in the behavior of the joint excitation function for ^{205}Em and ^{206}Em . (Fig. 7).

Francium-204 and Francium-204m. The two α peaks at 6.973 and 7.028 MeV (Fig. 3) have similar excitation functions as shown in Fig. 4. This suggests that they belong to the same isotope. The half-life of the 6.973 MeV peak was measured to be 3.3 ± 0.2 sec, that of the 7.028 MeV peak 2.2 ± 0.2 sec. This indicates that the peaks belong to two isomeric states of the same isotope. Griffioen and Macfarlane¹ have attributed the 7.028 MeV peak to ^{204}Fr on the basis of excitation functions and mass-energy systematics. Our observations are in agreement with this assignment. These authors made no statement concerning the 6.973 MeV peak although it does appear in their spectra. In Fig. 5 the excitation functions of the 6.412 and 6.464 MeV peaks of ^{200}At clearly follow those of the 6.973 and 7.028 MeV peaks in Fig. 4. In Fig. 7 the ^{204}Em excitation function resembles those of the 6.973 and 7.028 MeV peaks in Fig. 4.

On the basis of the systematic occurrence of isomerism in the light isotopes of emanation, astatine, and polonium, ^{204}Fr can be expected to occur in isomeric forms. We have assigned the 6.973 MeV peak to the ground state and the 7.028 MeV peak to an isomeric state in ^{204}Fr , as this fits best in the mass-energy systematics.

Francium-203. The α peak at 7.130 MeV with a half-life of 0.7 ± 0.3 sec is visible only in the spectra taken at the highest beam energies. Comparison of excitation functions indicates that the peak belongs to a francium isotope lighter than ^{204}Fr , most probably to ^{203}Fr . The α energy of ^{199}At , the α daughter of ^{203}Fr , is 6.638 MeV. In our measurements we could not resolve this α energy from the 6.647 MeV α group of ^{208}Fr . Hence, we attribute the sharp rise of the composite $^{209,208}\text{Fr}$ excitation curve at maximum beam energy in Fig. 4 to the presence of ^{199}At . The ^{203}Em excitation curve in Fig. 7 shows that ^{203}Fr is produced in the reaction. The α energy of 7.130 MeV is close to the value expected for ^{203}Fr from mass-energy systematics.

IV. DISCUSSION

This is one of a series of studies of nuclei above 82 in atomic number and below 126 in neutron number. Other reports have been prepared on the isotopes of emanation,² astatine,³ radium,⁴ and actinium.⁵

In the francium work, as in the studies of the other elements, the main reaction products appear to be made by compound nucleus reactions. The excitation functions for the francium isotopes have the expected appearance for compound nucleus reactions if account is taken of the fact that the ^{211}Fr - ^{210}Fr and ^{209}Fr - ^{208}Fr pairs have nearly identical α energies and hence that their excitation curves are composite. The experimental techniques were designed primarily for the study of the α -decay properties of the isotopes and were not suitable for quantitative measurements of cross sections. For the major products we could estimate relative production yields by correcting for

half-life and for electron-capture branching. According to the estimates of Griffioen and Macfarlane¹ this branching is less than 20% for all isotopes below mass 212. The general trend in the francium cross sections is a decrease with number of neutrons evaporated, which is reasonable on the grounds of fission competition at each stage of de-excitation.

The beam energy, E_{beam} , corresponding to the maximum yield for each francium isotope was used to determine the energy associated with the evaporation of each neutron. To accomplish this the Q value of the compound nucleus reaction was computed from the mass tables of Mattauch et al.¹⁶ and the neutron separation energies, S_i , were taken from the tables of Viola and Seaborg.¹⁷ The average energy dissipated per neutron evaporated, which is the sum of the kinetic energy of the neutron and the γ -ray energy, is given by the relation

$$\bar{E}_{\text{evap}} = \frac{E_{\text{beam}} + Q - \sum_{i=1}^n S_i}{n}$$

where n is the number of neutrons evaporated. In the $^{205}\text{Tl} + ^{12}\text{C}$ reaction the first four neutrons removed an average of 4.7 MeV and the next four an average of 4.5 MeV. In the $^{197}\text{Au} + ^{16}\text{O}$ reaction the first four evaporated neutrons removed an average of 5 MeV, and the next three an average of 4.1 MeV. These values are in agreement with the general result reported by Kaplan¹⁸ and by Alexander and Simonoff¹⁹ for heavy ion reactions with rare earth targets. They found that 4.8 to 6.3 MeV is dissipated for each neutron evaporated.

The case of ^{205}Fr requires a word of comment. The counting rate at the peak yield for the collected ^{205}Fr activity is manyfold less than expected. Most of the decrease can be attributed to decay of the 3.7 second isotope in

the time required to sweep it out of the reaction chamber. In our apparatus the effective mixing time within the chamber and sweep time through the orifice is sensitive to experimental conditions and difficult to measure exactly. We know it to be of the order of a few seconds so that decay in flight is no problem for activities with 10 sec or longer half lives. In future experiments we shall use a redesigned apparatus to increase the collection speed and thus permit better measurements of such isotopes as ^{205}Fr and ^{204}Fr . We cannot say whether the decreased yield is related to the anomolous position of the yield maximum for ^{205}Fr . This position corresponds to the dissipation of about 9 MeV in the evaporation of the last neutron, about twice the average for each of the preceding three neutrons.

Certain features of the excitation curves for the astatine isotopes (Fig. 5) are worthy of note. A large part of the astatine yield, particularly for those of higher mass, is the result of α decay of francium parent isotopes, which accounts for the appearance of yield maxima at the same bombarding energy as for the parent francium isotope. In addition there appears in several instances a more or less clearly defined second maximum at a beam energy about 20 MeV higher. We suggest that this second maximum is caused by direct reactions of the type $^{197}\text{Au}(^{16}\text{O}, \alpha, xn)^{209-x}\text{At}$. The 20 MeV displacement in the yield maximum probably has the significance that the instantaneous removal of an α particle requires that about 20 MeV of Coulombic energy be converted into kinetic energy of the α particle. It may also be noted that the emanation excitation curves show no evidence for a contribution by direct production, which in this case would have to come about by direct or indirect emission of protons or deuterons.

The α -decay energy of the francium isotopes as a function of neutron number follows the systematic trends below 126 neutrons observed for all elements from polonium through radium. This is discussed in a related article on the astatine isotopes³ and our data are included in Fig. 3 of that article.

Of special interest is the isomeric pair observed in ^{204}Fr , a nucleus with 117 neutrons. We have noted previously² that all odd neutron isotopes of the even Z elements, mercury, platinum, lead, polonium, and emanation with 117 or fewer neutrons exist in isomeric forms which can be associated with the $i_{13/2}$ shell model state. Similarly the odd-odd nuclei of astatine with 113, 115, and 117 neutrons have isomeric states³ which may be associated with a low-lying high-spin state formed by coupling the $i_{13/2}$ neutron with the odd proton. This ^{204}Fr isomerism can be explained in a similar way.

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FOOTNOTES AND REFERENCES

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Table I. Present results compared with those of Griffioen and Macfarlane.¹
 The following energy standards were used: ²¹²Po, 8.7854 MeV; ²¹⁵Po, 7.3841 MeV; ²¹⁹Em, 6.8176 MeV; ²¹¹Bi, 6.6222 MeV; ²¹²Bi, 6.0898 MeV; ²¹²Bi, 6.0506 MeV; ²¹⁶Po, 6.7772 MeV; ²²⁰Em, 6.2884 MeV; ²²⁴Ra, 5.6840 MeV.¹²

Isotope	This work		Griffioen and Macfarlane	
	Alpha energy (MeV)	Half-life	Alpha energy (MeV)	Half-life
²⁰³ Fr	7.130±0.005	0.7 ± 0.3 sec		
^{204m} Fr	7.028±0.005	2.2 ± 0.2 sec	7.02±0.03	2.0±0.5 sec
²⁰⁴ Fr	6.973±0.005	3.3 ± 0.2 sec		
²⁰⁵ Fr	6.917±0.005	3.7 ± 0.2 sec	6.91±0.02	3.7±0.4 sec
²⁰⁶ Fr	6.792±0.005	15.7 ± 0.3 sec	6.79±0.02	15.8±0.4 sec
²⁰⁷ Fr	6.773±0.005	14.7 ± 0.3 sec	6.77±0.02	18.7±0.8 sec
²⁰⁸ Fr	6.647±0.005	59 ± 2.0 sec	6.65±0.02	37.5±2.0 sec
²⁰⁹ Fr	6.647±0.005	52 ± 2.0 sec	6.65±0.02	54.7±1.0 sec
²¹⁰ Fr	6.537±0.005	3.18±0.06 min	6.55±0.02	2.65±0.08 min
²¹¹ Fr	6.533±0.005	3.06±0.06 min	6.55±0.02	3.10±0.07 min
²¹³ Fr	6.773±0.005	34.7 ± 0.3 sec	6.77±0.01	33.7±1.5 sec
²¹³ Em	8.090±0.008		8.13 ^a	

^aRef. 13.

FIGURE CAPTIONS

Fig. 1. Alpha spectra showing francium and daughter activities produced in the reactions $^{205}\text{Tl}(^{12}\text{C},\text{xn})^{217-\text{x}}\text{Fr}$ at three beam energies. Recoil collection and counting cycle periods were 0.8 sec and the total measuring time 7 min. The beam current was 0.05 μA .

Fig. 2. Excitation functions of the francium and daughter activities produced in the reactions $^{205}\text{Tl}(^{12}\text{C},\text{xn})^{217-\text{x}}\text{Fr}$. These curves were derived from spectra such as those shown in Fig. 1, taken successively from high to low beam energies.

Fig. 3. Alpha spectra showing francium and daughter activities produced in the reactions $^{197}\text{Au}(^{16}\text{O},\text{xn})^{213-\text{x}}\text{Fr}$ at three beam energies. The beam current was 0.5 μA . Recoil collection and counting cycle periods were 0.8 sec. Total measuring time was 7 min. The inserted spectrum shows ^{203}Fr in a longer measurement at 161 MeV beam energy.

Fig. 4. Excitation functions for the francium isotopes produced in the reactions $^{197}\text{Au}(^{16}\text{O},\text{xn})^{213-\text{x}}\text{Fr}$. These curves were derived from spectra similar to those in Fig. 3, taken successively from low to high beam energies. Excitation functions for the daughter activities observed in the same measurements are shown in Fig. 5.

Fig. 5. Excitation functions for astatine isotopes and $^{207,208}\text{Em}$ produced in the reactions of ^{197}Au with ^{16}O . The curves for long-lived isotopes are somewhat distorted toward higher energy owing to persistence of activity from one measurement to the next. Compare with the francium curves in Fig. 4.

Fig. 6. Alpha spectra showing emanation activities produced in the reactions of ^{197}Au with ^{16}O at two beam energies. Measuring times were 40 min. Beam current was $1.2\ \mu\text{A}$.

Fig. 7. Excitation functions of the emanation isotopes produced in the reactions of ^{197}Au with ^{16}O . The curves were run from high to low beam energies. Measuring times were 40 min and the intervals between them 3 min. Beam current was $1.2\ \mu\text{A}$.

XBL672-879

Fig. 1.

XBL672-680

Fig. 2.

Fig. 3.

XBL 671-184

XBL671-183

Fig. 4.

XBL672-88I

Fig. 5.

XBL672-882

Fig. 6.

XBL672-883

Fig. 7.

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