unaffected by prolonged exposure. The 0.1N HCl which is the usual internal reference solution of a glass electrode is relatively stable (6), and neutral alkali chloride solutions are unaffected. Thus, one criterion for the behavior of an electrode after large dose is the stability of its internal reference solution.

Solid state changes may also affect the performance of an electrode. A good example here is the asymmetry potential of the glass electrode which undergoes minor but permanent changes. It is possible that the europium-doped lanthanum fluoride single crystal which is the sensing unit of the fluoride electrode also undergoes some change in internal structure.

Stability of the Glass and Saturated Calomel Electrode to Alpha Radiation. The glass and calomel electrodes were also tested for their stability to alpha radiation. The radiation source was ^{244}Cm in 3M HCl that was prepared from the metal oxide, and its activity was close to 10^9 dps/ml. No dose rate measurement was made, but the dose rate calculated from

the disintegration rate was of the order of 10^4 rad/min. Microelectrodes were used to keep down the volume of the alpha solution required. Since this solution was kept acidic to retain the curium in solution, each electrode was removed from the hot solution, and pH readings were taken in pH 4, 7, and 10 buffers using a counter electrode that was not irradiated. The counter electrodes were checked against each other before each set of measurements. Readings taken at various intervals over a two-week period (cumulative dose close to 10^8 rad) showed no significant change.

From the experience with gamma radiation, little effect to these electrodes was expected from pure alpha emitters. It is felt that adverse effects are more likely only when there is relatively energetic gamma associated with the alpha decay; the type of effects described for gamma radiation then are expected to take place.

(6) H. Kubota, ANAL. CHEM., **40**, 271 (1968).

Sources and Prevention of Recoil Contamination of Solid-State Alpha Detectors

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Most recoil contamination of the solid-state detectors used in alpha spectrometry results from radioactive chains in which short-lived, alpha-emitting daughters are produced either directly from alpha-emitting parents or indirectly by decay of a short-lived beta-emitting daughter which in turn was produced from an alpha-emitting parent. Spectra are given showing the recoil products resulting from various combinations of exposure and decay time for each of the four main radioactive series. Recoil contamination of the detector can be reduced by a factor of at least 10^3 with a loss in resolution of only 1 or 2 keV by leaving enough air in the counting chamber to produce 12 μg/cm² of absorber between the source and detector, and applying a negative potential of 6 volts to the source plate. Polonium-210 gives a marked pseudo-recoil effect, probably due to its own inherent volatility, and is by far the most serious long-term contaminant encountered. Its spontaneous volatilization from a stainless steel support can be virtually eliminated by heating the plate for 5 minutes at about 300 °C without significant loss of resolution.

BECAUSE OF THEIR HIGH RESOLUTION and low backgrounds, solid-state surface-barrier detectors permit exceptionally high sensitivity and reliability in the detection and identification of alpha-emitting radionuclides by alpha spectrometry. However, the detectors are expensive, particularly in the larger sizes, and are susceptible to contamination during use that is virtually impossible to remove without damaging the detector. Consequently, both sensitivity and reliability decrease with use. The present work was carried out in an attempt to identify the most common sources of contamination and to find simple means of minimizing their effect.

Emission of a particle during radioactive decay results in recoil of the residual nucleus in the opposite direction because

of the fundamental requirement that momentum be conserved in each individual radioactive disintegration. Hahn and Meitner (1) were among the first to call attention to the usefulness of recoil collection in radiochemistry. Since then, the recoil technique has been used extensively for preparing extremely thin sources for alpha spectrometry (2), for making rapid radiochemical separations (3), and in "hot atom" chemistry in general (4). The technique has been very useful in establishing genetic relationships among the members of both the natural radioactive series (5) and the new collateral series resulting from deuteron bombardment of thorium (6, 7), particularly in determining the half-lives of the short-lived members.

Emission of a beta particle produces too little recoil energy to cause significant contamination under usual conditions. However, emission of an alpha particle can result in contamination of the detector due to the accompanying recoil atoms. A 5-MeV alpha particle produces recoil energy of about 100 keV in a residual atom of mass 200, or about 2%

(1) O. Hahn and L. Meitner, *Phys. Z.*, **10**, 697 (1909).
(2) B. G. Harvey, "Introduction to Nuclear Physics and Chemistry," Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1962, p 318.
(3) Y. Kusaka and W. W. Meinke, "Rapid Radiochemical Separations," *Nat. Acad. Sci.–Nat. Res. Counc., Nucl. Sci. Ser.* NAS-NS 3104 (1961).
(4) A. C. Wahl and N. A. Bonner, "Radioactivity Applied to Chemistry," Wiley, New York, N. Y., 1951, Chapter 8.
(5) C. M. Lederer, J. M. Hollander, and I. Perlman, "Table of Isotopes," Sixth Ed., Wiley, New York, N. Y., 1967.
(6) M. H. Studier and E. K. Hyde, *Phys. Rev.*, **74**, 591 (1948).
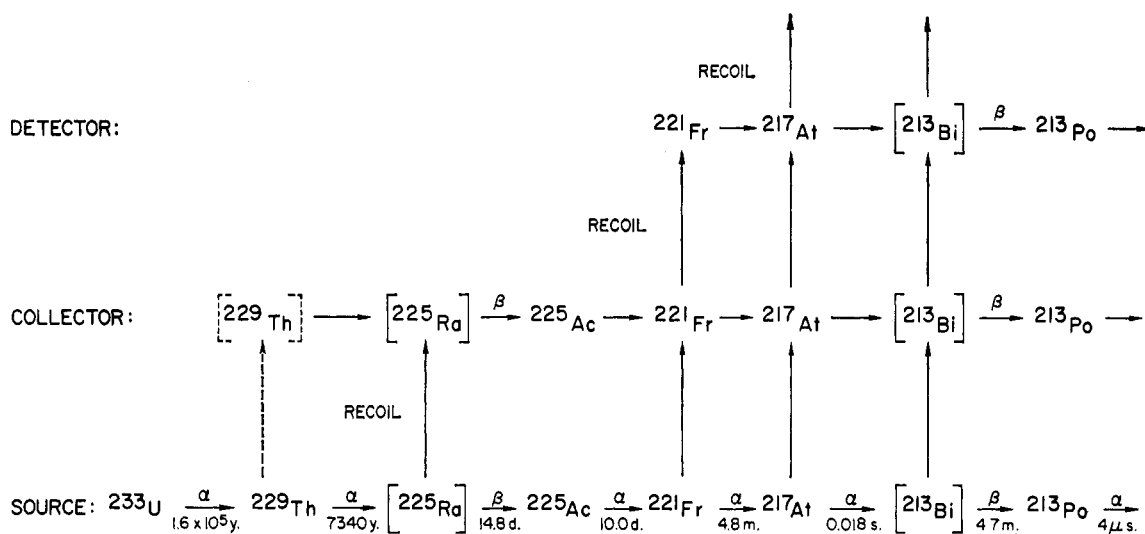(7) W. W. Meinke, A. Ghiorso, and G. T. Seaborg, *ibid.*, **81**, 782 (1951).

Figure 1. Recoil atoms from the ^{233}U chain

of the kinetic energy of the alpha ray producing the recoil. This energy is sufficient to produce an average range in air of about 0.1 to 0.2 mm at atmospheric pressure (8). At the reduced pressures and short source-to-detector distances used in alpha spectrometry, the detector will collect almost as many recoiling atoms as alpha particles because the geometry is the same for both.

Most long-lived alpha emitters present no problem from the standpoint of recoil because they decay to daughters that are either long-lived or beta emitters. Most cases of contamination by recoil undoubtedly result from radioactive chains in which short-lived, alpha-emitting daughters are produced either directly from alpha-emitting parents or by decay of a short-lived beta-emitting daughter which in turn was produced from an alpha-emitting parent. A typical example is shown by the decay chain of ^{233}U in Figure 1. Each collector will act in turn as a source for further recoil of succeeding daughters to a secondary collector—e.g., the detector used to count the primary collector. Because the recoil process takes place on an atom-by-atom basis, the extent and severity of the contamination in terms of activity depend on the half-lives of the recoil daughters and their progeny. The highest immediate contamination will result from short-lived daughters with half-lives of less than a few hours but is not too serious because the activity will decay in an acceptably short period of time such as overnight. The most serious contamination will likely result from daughters with half-lives of a few days or weeks which are short enough to result in a build-up of appreciable activity during reasonable counting times but long enough to require an untenably long time for the contaminant to decay. Daughter atoms with half-lives longer than a few months can be prevented from accumulating in the large numbers required to produce significant activity by good management. Obviously, direct contamination by long-lived alpha emitters cannot be tolerated.

For a given number of recoil atoms, the activity obviously decreases with increasing half-life. For example, during decay of ^{233}U, ^{229}Th recoils like the shorter-lived daughters but the activity produced during any reasonable exposure

is very small because of its long half-life. If the detector collects the recoil atoms as efficiently as the alpha particles, a 10^4-cpm source of ^{233}U counted for 1 minute each day for 1 year would give only 1 disintegration per day of contamination from its ^{229}Th daughter. In contrast, an ^{225}Ac source of the same strength would result in contamination of 1.5×10^3 dpm of its 4.8-min ^{221}Fr daughter from a single 1-min count.

In the present investigation, alpha spectra of all recoil products resulting from each of the four long-lived radioactive series were obtained with the large, high-resolution, solid-state detectors that have become available in the last few years. The actual sources used were ^{226}Ra, ^{227}Ac, ^{228}Th, and ^{229}Th which are the immediate long-lived precursors of all the daughters capable of giving significant recoil activity in a reasonable length of time. The spectra obtained with different exposure and decay times are interpreted in terms of the well-known genetic relationships existing among the various members of each chain. Similar recoil contaminants will result from the collateral series but will be encountered only in bombardment work and are not included here. In most cases, the half-lives of the recoil daughters are so short as to produce little lasting contamination.

EXPERIMENTAL

Instrumentation. Most of the alpha energy spectra were obtained with a 450-mm^2 surface-barrier detector, a charge-sensitive preamplifier, linear amplifier, biased amplifier, pulse stretcher, and a 400-channel pulse-height analyzer. The resolution obtained with a ^{210}Po source electrodeposited on polished platinum was 22 keV full width at half maximum peak height (FWHM). The counting efficiency for a 1-inch source on the top shelf (ca. 0.64 cm) was 25.1%. Where noted, a 50-mm^2 detector was used having a resolution of about 14 keV FWHM.

Sources. The ^{226}Ra source was prepared by direct evaporation of a carrier-free solution onto a 2-inch mirror-finished stainless steel plate in a central area about 1 inch in diameter. The plate was then ignited briefly to minimize absorbing solids. All other sources were electrodeposited onto identical stainless steel disks over a similar area.

Procedures. In determining the effect of absorber on the resolution and apparent energy of ^{210}Po, the regular chamber of the alpha spectrometer was used. However, when sources having recoilable daughters were being measured, other

(8) E. Rutherford, J. Chadwick, and C. D. Ellis, "Radiations from Radioactive Substances," Cambridge University Press, London, 1930, p 155.
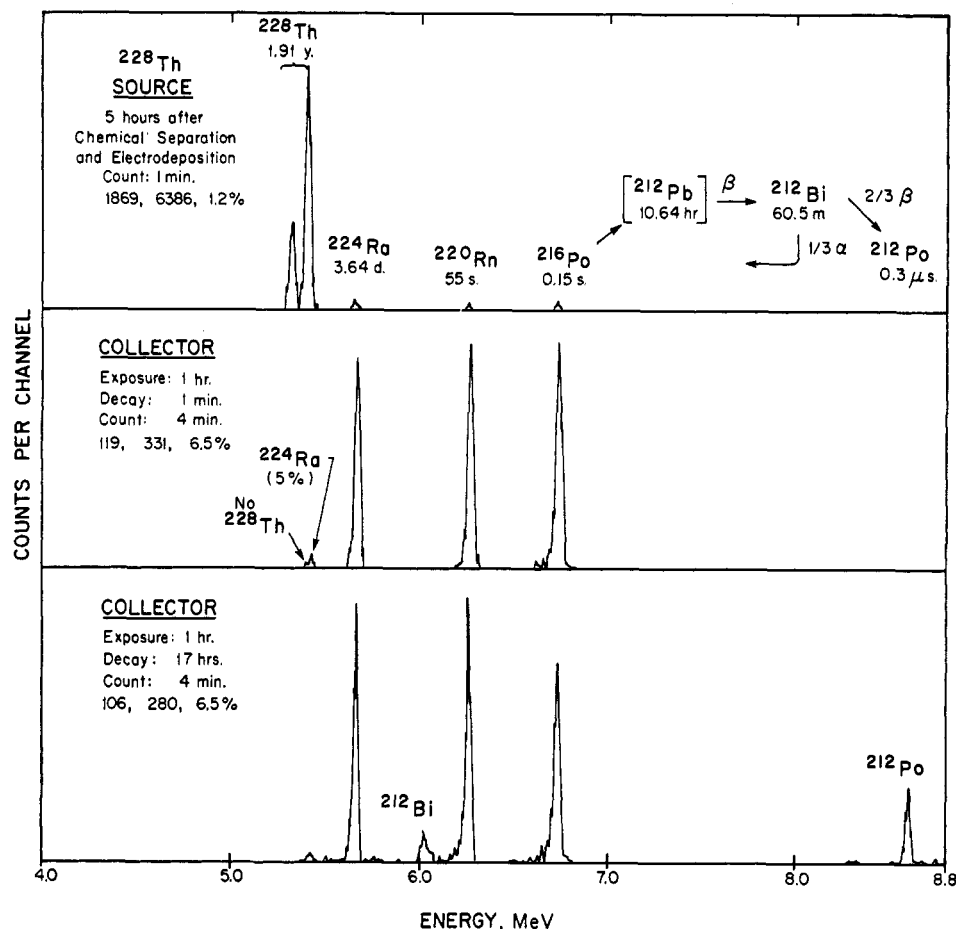
Figure 2. Recoil atoms from freshly-purified ^{228}Th without daughters

chambers were used to avoid contamination of the detector with the longer-lived daughters. For identification of the recoil products, the recoiling atoms were collected on 2-inch mirror-finished stainless steel disks separated from the source by a 1-mm plastic spacer ring in a vacuum desiccator at about 0.1-mm pressure. At the end of the exposure, the collector plate was transferred to the alpha spectrometer and a spectrum of the recoil activity was obtained. A blank plate was then substituted for the collector plate, and a background spectrum was obtained to determine secondary recoil contamination of the detector from the primary collector plate.

To determine the effect of pressure on recoil contamination, a freshly-purified and electrodeposited ^{225}Ac source was employed to provide a high recoil activity in a short collection period from the 4.8-minute ^{221}Fr daughter. The source and stainless steel collector plate were placed in a chamber with a removable barrier so that recoiling atoms could not strike the collector plate until the collecting period was begun. The chamber was evacuated to the desired pressure, the barrier was removed, and the recoiling atoms were collected for 5 minutes. The collector plate was then transferred quickly to the spectrometer chamber to eliminate counting alphas embedded in the walls of the collection chamber, and a spectrum of the recoil activity was obtained at <1-mm pressure. The time from the end of exposure to the beginning of the counting period was kept constant at 2 minutes to provide for transfer and evacuation and the counting time was kept constant at 4 minutes. The area under the ^{221}Fr peak was used to evaluate the effect of pressure on the number of recoils obtained.

RESULTS AND DISCUSSION

The composition of the recoil activity obtained is presented in relation to the composition of the source, the length of the exposure, the decay time before counting, and the counting time. The multiple spectra in each figure have been aligned vertically for ease of identification of the nuclides present and their subsequent growth and decay relationships. A semi-quantitative idea of the relative activities of both source and recoil plates and the statistical uncertainties involved in the measurements can be obtained from the three numbers given at the bottom of the legend in each spectrum. These numbers refer, respectively, to the number of counts in the peak channel and the total number of counts under the entire peak for the nuclide having the highest peak, i.e., the ordinate and area, and the counting efficiency for the particular shelf used in counting. For example, the small ^{224}Ra peak in the top spectrum of Figure 2 contained 76 cpm in the peak channel and a total of 214 cpm under the peak. When corrected for the difference in counting time and efficiency, this peak is actually 13 times higher and larger than that shown in the middle spectrum.

Thorium-228. Figure 2 shows the recoil obtained from ^{228}Th shortly after its chemical separation from aged ^{228}Ra when the ^{228}Th daughter activity is very low. When the exposure time is short, the initial recoil activity is composed entirely of ^{224}Ra and its two immediate short-lived daughters. After several hours' decay, ingrowth of ^{212}Bi and ^{212}Po through the 10.64-hour ^{212}Pb intermediate becomes appreciable as shown in the bottom spectrum. Significantly, no ^{228}Th could be detected, proving conclusively that the daughter atoms on the collector are indeed produced by recoil rather than by some other inadvertent mechanical contamination. The small peak at the ^{228}Th energy is a 5% branch of ^{224}Ra.
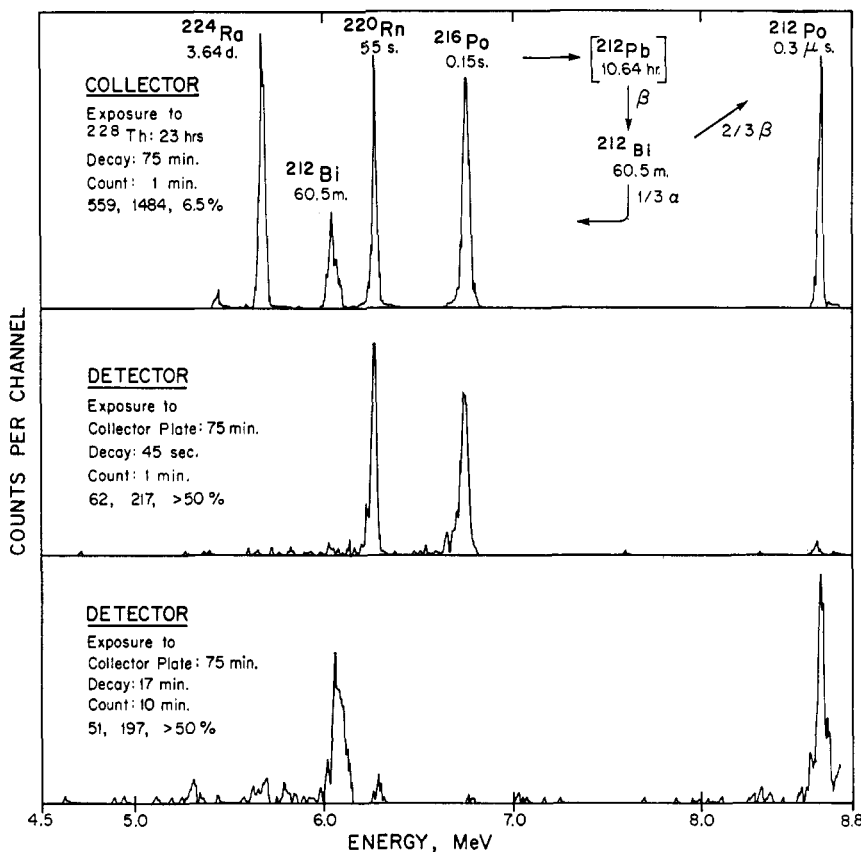
Figure 3. Recoil atoms from ^{228}Th containing daughters

As shown in the top spectrum of Figure 3, the sum of the ^{212}Bi and ^{212}Po activities (which result from branching decay and must be summed to be equivalent to the other nuclides) soon exceeds that of the 3.64-day ^{224}Ra because of the shorter half-life of the ^{212}Pb parent despite the larger number of ^{224}Ra recoils from the ^{228}Th parent. When the ^{228}Th source is itself old enough to contain near-equilibrium concentrations of its daughters, even the initial concentration of ^{212}Bi plus ^{212}Po from short exposures will be many times that of the ^{224}Ra. (See Figure 6 below.) For the same number of atoms collected on the recoil plate, the theoretical ratio of the activity of 10.64-hour ^{212}Pb to that of 3.64-day ^{224}Ra obtained in 1 hour is 7.9. The ^{220}Rn and ^{216}Po activities seldom exceed that of their parent because their half-lives are so short that the excess activity will have decayed before the count is made. In fact, the short-lived ^{220}Rn and ^{216}Po daughters generally show progressively smaller activities than that of ^{224}Ra because of recoil loss from the collector during the time that the plate was in the evacuated chamber. Each atom lost not only reduces the number of atoms remaining of all succeeding nuclides but also produces a greater decrease in activity of the nuclides with the shorter half-lives. No trace of ^{228}Th could be detected with even the longest exposures.
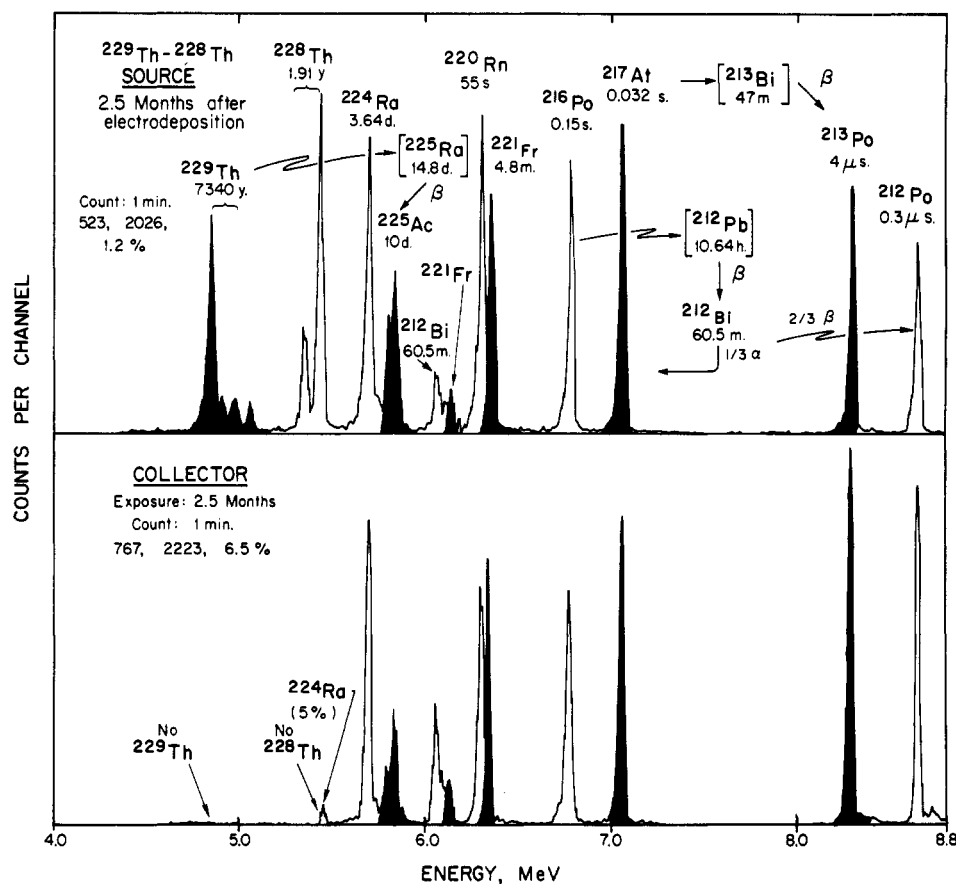
The middle spectrum in Figure 3 shows the successive contamination of the detector from counting the primary collector plate to be essentially ^{220}Rn from recoil accompanying decay of its immediate parent ^{224}Ra on the collector plate. The ^{216}Po present cannot exceed that which can be supported by the ^{220}Rn on the detector (less some recoil loss) because of the decay of any unsupported activity during the exposure. After several half-lives of the 55-sec ^{220}Rn, the only activity remaining on the detector is that from ^{212}Bi and

^{212}Po supported by the 10.64-hour ^{212}Pb as shown in the bottom spectrum. Because of its frequent occurrence from decay of both naturally occurring ^{232}Th and synthetic ^{232}U, and the 3.64-day half-life of its first recoilable daughter, ^{228}Th is undoubtedly one of the most important sources of relatively long-lived contamination to be encountered.

Thorium-229. The shaded curve of Figure 4 shows the ^{229}Th chain outlined in Figure 1 about 2.5 months after its separation from several milligrams of old ^{233}U. Exposure of the collector plate was begun shortly after preparation of the source to permit considerable ingrowth of ^{225}Ac following ingrowth and recoil of ^{225}Ra which does not electrodeposit. The open curve shows the ^{228}Th chain resulting from decay of ^{232}U impurity in the ^{233}U. The collector plate shows the same characteristics for both series as described above. Neither parent of either series is detectable despite the long exposure. The activities of ^{220}Rn and ^{216}Po on the collector are approximately equal to each other because of their short half-lives but markedly less than that of ^{224}Ra because of recoil loss. The activities of ^{221}Fr and ^{217}At are also equal to each other, when the minor branch of ^{221}Fr is included, but greater than the ^{225}Ac activity which has not yet grown to equilibrium. Both ^{213}Po and the sum of the ^{212}Po plus ^{212}Bi branches are significantly higher than any of their precursors. Because this effect in the ^{228}Th chain has been erroneously attributed by others to outgassing of radon, the same result with ^{213}Po from the ^{229}Th chain, which does not contain a radon isotope, proves conclusively that a gaseous intermediate is not necessary.

Figures 5–7 show the recoil atoms obtained with much shorter exposure to this same aged mixture of ^{228}Th and ^{229}Th containing near-equilibrium concentrations of daughters. With an exposure time of 10 minutes (Figure 5), only

Figure 4. Recoil atoms from long exposure to mixture of ^{228}Th and ^{229}Th

229Th-228Th SOURCE
2.5 Months after electrodeposition
Count: 1 min.
523, 2026, 1.2 %

COLLECTOR
Exposure: 2.5 Months
Count: 1 min.
767, 2223, 6.5 %

ENERGY, MeV

COUNTS PER CHANNEL

the short-lived progeny are present shortly after the exposure. Neither ^{224}Ra nor ^{225}Ac have built up to detectable levels. The bottom two spectra show the decay of the 55-sec ^{220}Rn and its daughter followed later by the decay of 4.8-min ^{221}Fr and its daughter with an increase in the relative activity of ^{213}Po which is supported by 47-min ^{213}Bi. The ^{213}Po then decays with a 47-min half-life.

After several hours' exposure (Figure 6), the activity of ^{224}Ra and its daughters becomes significant. Again, the activities of ^{213}Po and the sum of ^{212}Bi and ^{212}Po are much larger than any of their respective precursors. The ^{221}Fr and ^{217}At decay completely in a few minutes and the ^{213}Po in a few hours, leaving only the longer-lived ^{224}Ra series with a heavily disproportionate activity of ^{212}Po plus ^{212}Bi. After several more hours' decay (Figure 7), the excess ^{212}Po and ^{212}Bi activities, supported only by the 10.64-hour ^{212}Pb, have decayed significantly relative to the other activities. However, note the marked decrease in activity from ^{224}Ra to ^{220}Rn to ^{216}Po due to recoil loss from the collector plate. After 4 days, the excess activity has decayed nearly back to equilibrium with the ^{216}Po but the sequential drop in activity from ^{224}Ra to ^{216}Po is still readily apparent. The most serious long-term contamination resulting from counting ^{229}Th will not be apparent immediately because of the time required for the 10-day ^{225}Ac to grow in from the beta-emitting ^{225}Ra, the actual recoil contaminant. The middle spectrum of Figure 7 shows the ^{225}Ac and its francium, astatine, and polonium daughters in equilibrium concentrations that have grown in.

The bottom spectrum of Figure 7 shows that the secondary recoil contamination on the detector shortly following the spectrum made of the primary collector plate begins with the first recoilable daughter produced by alpha-emission of

the parent in the source. Besides being an interesting spectrum of a very short-lived gaseous nuclide and its daughter unsupported by longer-lived parents of lower inherent volatility, as was also found in the middle spectrum of Figure 3, this spectrum also shows the small quantities of longer-lived nuclides built up on the detector during the course of this investigation. Even though the small blips represent only 1 and 2 pulses, they always occur at the exact energies corresponding to the other nuclides present in both the ^{228}Th and ^{229}Th series, showing the sensitivity and certainty with which extremely minute levels of activity can be identified by alpha spectrometry on a clean detector.

Radium-226. The top spectrum of Figure 8 shows the ^{226}Ra source containing very small quantities of ^{222}Rn and ^{218}Po that have grown in during the 5 hours allowed for decay of radon daughters from the original solution. The ^{210}Po is residual activity unvolatilized from the original solution and should be ignored. The recoil activity is predominately 3.05-min ^{218}Po resulting from that produced by direct recoil from the source plus that resulting from decay of the ^{222}Rn on the collector plate. (The ^{222}Rn activity on the source was 97 cpm at 1.2% counting efficiency compared to 87 counts in 4 minutes at 21.3% for the ^{218}Po on the collector.) The ^{214}Po is the ingrowth through the 30-min ^{214}Pb–^{214}Bi intermediate during the time of exposure. With longer exposures, the ^{214}Po grows in to higher levels than its precursors as was found with the previous examples. Again, the ^{218}Po activity on the collector plate is distinctly lower than that of its immediate precursor when the time in the evacuated chamber is long compared to its 3-min half-life.

Figure 9 shows the recoil atoms obtained from a ^{226}Ra containing large quantities of ^{222}Rn and its short-lived daughters. As with the other sources, no ^{226}Ra parent could be
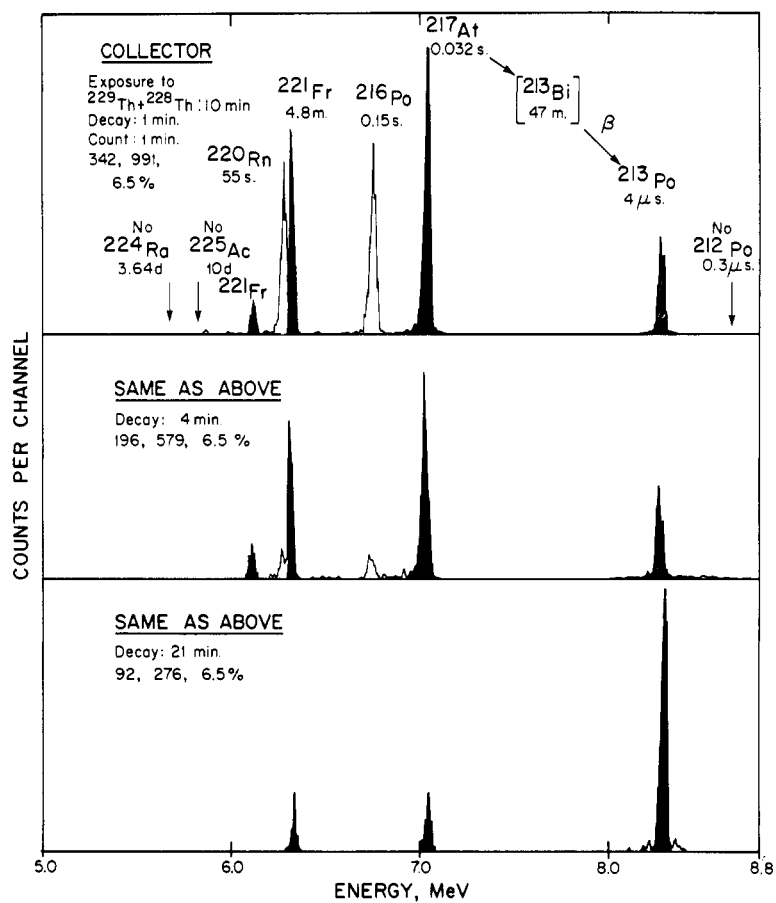
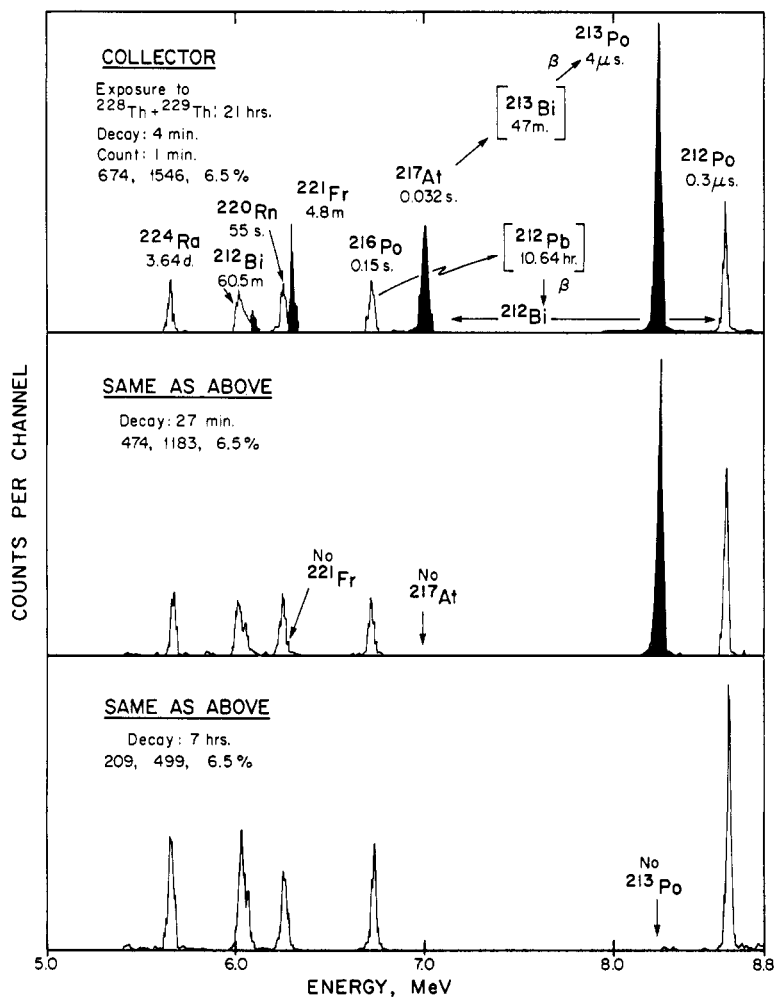Figure 5. Recoil atoms from short exposure to aged mixture of ^{228}Th and ^{229}Th

Figure 6. Recoil atoms from medium exposure to aged mixture of ^{228}Th and ^{229}Th after short decay

Figure 7. Recoil atoms from medium exposure to aged mixture of ^{228}Th and ^{229}Th after long decay

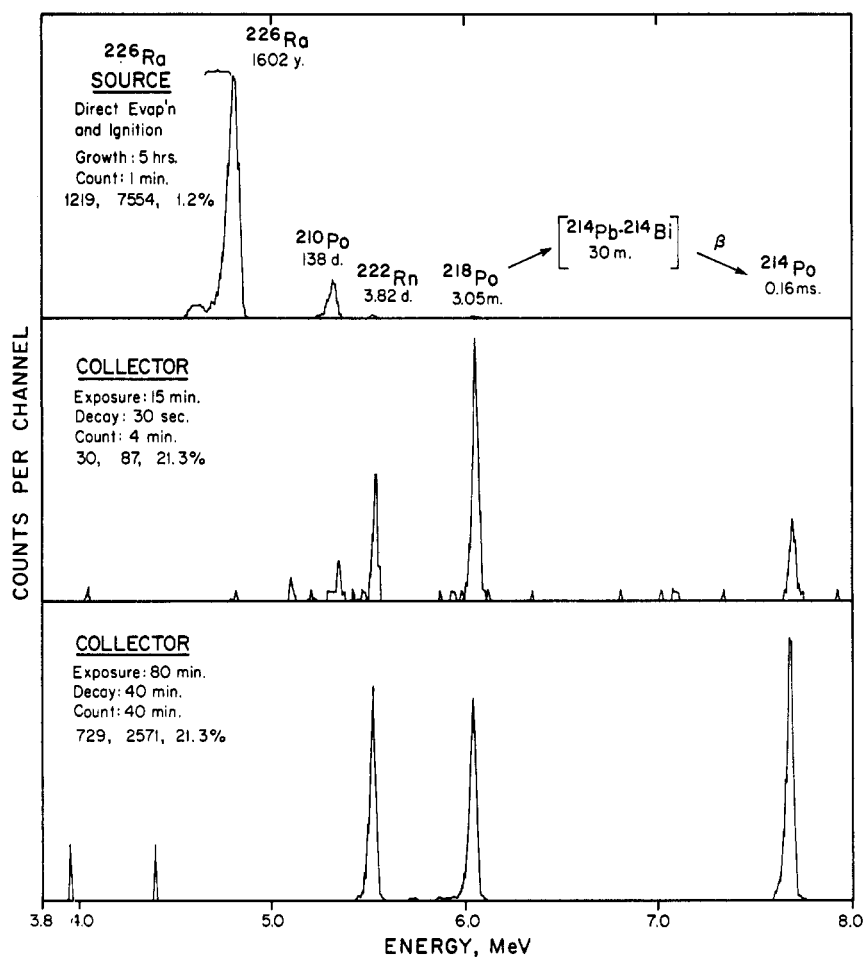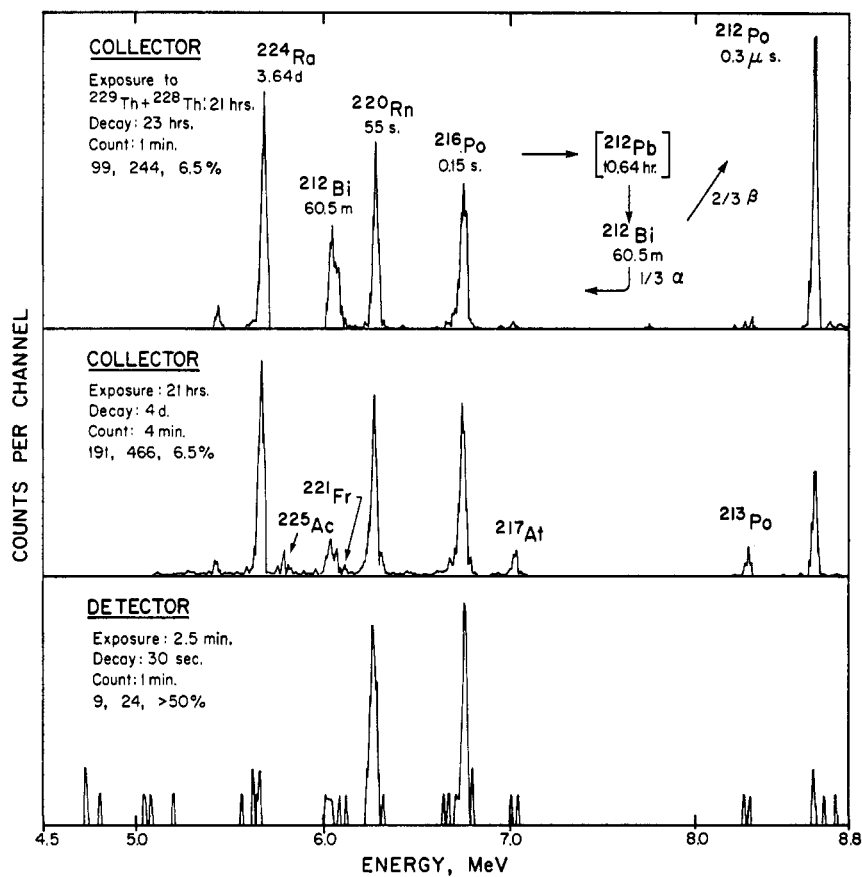Figure 8. Recoil atoms from ^{226}Ra without daughters

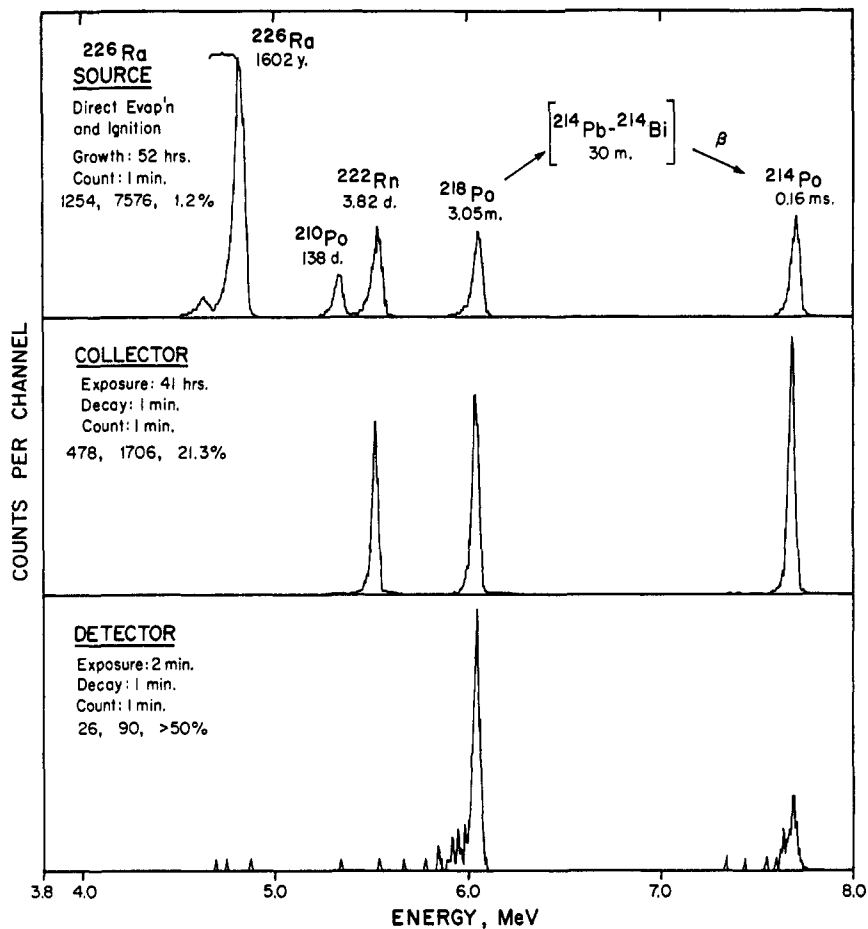**Figure 9. Recoil atoms from ²²⁶Ra containing daughters**

detected on the collector plate. More ^{218}Po is present than ^{222}Rn because the spectrum was obtained so quickly after removal of the source that the excess ^{218}Po present from direct recoil has not decayed to its equilibrium position with the ^{222}Rn on the collector plate. The bottom spectrum shows that the contamination on the detector is essentially ^{218}Po with a smaller quantity of ^{214}Po and no detectable ^{222}Rn. Besides its interest in showing a 3-min emitter unsupported by a longer-lived parent, this spectrum demonstrates again that only the residual nucleus accompanying an alpha decay will recoil without being accompanied by the parent undergoing the disintegration, in this case ^{222}Rn. After a few more minutes, the ^{218}Po decays and the detector contamination consists entirely of the 30-min ^{214}Po.

Neither the ^{222}Rn nor ^{220}Rn on the above collector plates displayed any tendency to disappear from the plate faster than required by radioactive decay. Although the recoil energy might be expected to bury the recoiling atom deeply enough in the steel plate to prevent its easy removal, other experiments were performed to determine how difficult removal of gaseous atoms might be. A polished stainless steel plate was exposed to the ^{226}Ra source in a vacuum for 8 days. A 40-minute spectrum of the recoil plate was similar to the middle one of Figure 9 and contained 1.33 × 10⁵ total counts under the ^{222}Rn peak. The collector plate was then placed in a vacuum desiccator at a pressure of 0.1 mm for 20.5 hours and another 40-minute spectrum was obtained. After correcting for decay, the before-and-after counts were identical within the statistical uncertainty of 0.8% at the 95% confidence level, showing that the gaseous activity does not dissipate detectably over long periods of time in a vacuum.

The same plate was then heated to a bright cherry red color over a blast lamp for 1 minute and another 40-minute spectrum was obtained. The ^{222}Rn activity decreased by only 3.6 ± 0.4%. Another measurement made on a different plate gave the same loss in 30 seconds but at a somewhat higher temperature. Radon atoms collected by recoil in a vacuum at close range are indeed held most tenaciously on a stainless steel plate. Consequently, recoil collection is a very simple and convenient method for preparing sources of radon isotopes for alpha spectrometry. Most of the contamination from ^{222}Rn and its daughters resulting from counting sources containing ^{226}Ra is almost certainly to have been caused by recoil rather than by outgassing of the ^{222}Rn as is frequently claimed.

Actinium-227. Because of the multiple branching of both ^{227}Th and ^{223}Ra and the similarity in the energy of several of their major branches, the recoil characteristics of the ^{227}Ac daughters are a little more difficult to display than the previous examples in which the alpha energies are well separated. The top spectrum of Figure 10 shows the ^{227}Ac source shortly after electrodeposition. Because the ^{227}Ac itself is primarily a beta-emitter, and the ^{223}Ra neither electrodeposits nor grows back rapidly due to its 11.4-day half-life, the spectrum of ^{227}Th is shown clearly along with the ^{211}Bi from the original solution and a very small quantity of ^{219}Rn and ^{215}Po (undoubtedly supported by an equally small quantity of ^{223}Ra). The solid curve of the middle section shows the decay of the original ^{211}Bi, supported by its 36-min ^{211}Pb precursor, in 3 hours. The open curve shows the significant ingrowth of ^{223}Ra and its radon, polonium, and bismuth daughters that occurred in 3 days. The top spectrum of Figure 11 shows how
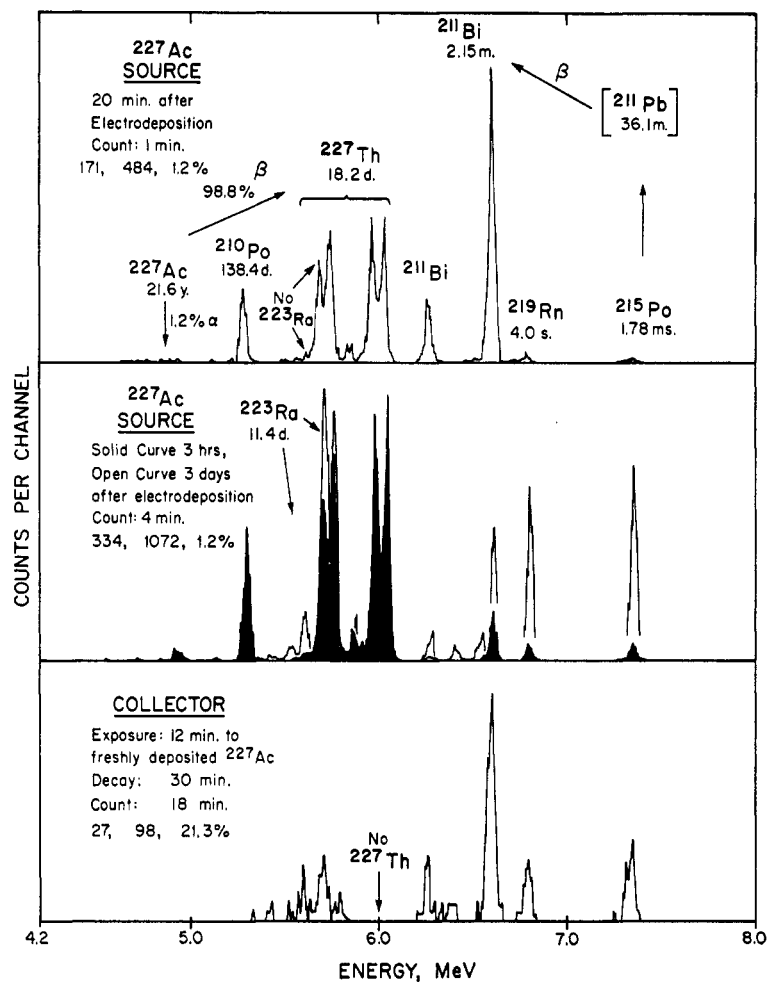
Figure 10. Recoil atoms from freshly electrodeposited ^{227}Ac

further ingrowth of ^{223}Ra almost obliterates the fine structure of the left side of the ^{227}Th spectrum. The spectrum of ^{223}Ra itself is shown clearly in the middle spectrum of Figure 11.

The recoil atoms resulting from short exposure to freshly electrodeposited ^{227}Ac are shown in the bottom spectrum of Figure 10. Even with a 30-minute decay to decrease the ^{211}Bi activity for clearer presentation of the other activities, the recoil activity is still mainly ^{211}Bi from decay of 36-min ^{211}Pb resulting from recoil as expected. However, significant recoil of the longer-lived ^{223}Ra and its two short-lived daughters from ^{227}Th is clearly evident with long counting times. The recoil obtained on long exposure to an aged ^{227}Ac source is shown in the middle part of Figure 11. The spectrum shows the relatively larger quantity of ^{211}Bi still present because of the short decay between exposure and counting and the significant ingrowth of ^{223}Ra and its daughters in fairly pure form. Because of lack of significant recoil accompanying the beta-emission from ^{227}Ac, no ^{227}Th is detectable on either collector plate. The only contamination remaining on the detector after having counted the collector plate is ^{211}Bi as expected. Because ^{227}Th was not present on the collector plate, which is the source plate for recoil to the detector, only the daughters of ^{223}Ra can recoil. Of these, the ^{219}Rn and ^{215}Po will have decayed off before the spectrum was obtained, leaving only the ^{211}Bi supported by the 36-min ^{211}Pb.

Polonium-210. Although no substantial recoil is possible, ^{210}Po gives a marked pseudo recoil effect, probably due to its own inherent volatility, and is by far the most serious contaminant encountered in the present investigation. When electrodeposited onto stainless steel under conditions used for deposition of electropositive elements as hydroxides, ^{210}Po activity transfers in significant quantities to another plate under the same conditions used for measuring recoil. A source containing 4.4×10^4 cpm of ^{210}Po contaminated an identical steel plate at a distance of 1 mm and a pressure of 0.1 mm of mercury to the extent of 106 cpm in 16.5 hours for an average rate of 6.4 cpm per hour of exposure when freshly prepared, or 1.5×10^{-4} cpm per hour of exposure per cpm of source activity. A few per cent of the collected activity disappeared within a few hours after which the remainder decayed with essentially the proper half-life, showing that some of the activity is very loosely held. After standing at atmospheric pressure for 13 days, the same source contaminated another plate to the extent of only 0.33 cpm per hour of exposure, a 20-fold decrease in apparent volatility. Another run with 3.8×10^4 cpm gave about 3 cpm per hour of exposure when freshly prepared and 180-fold less 16 days later. When the exposure was made at atmospheric pressure, the volatility was less than 0.01 cpm per hour of exposure, even when freshly prepared. The volatility is markedly dependent on the exact conditions of electrodeposition because other samples prepared under supposedly identical conditions showed very little volatility in a vacuum. In contrast, a ^{239}Pu source containing 1.5×10^5 cpm gave no contamination detectable in a 1-hour count from a 91-hour exposure in a vacuum, or $<3 \times 10^{-9}$ cpm per hour of exposure per cpm of source activity due to "aggregate" recoil from the alpha activity itself.
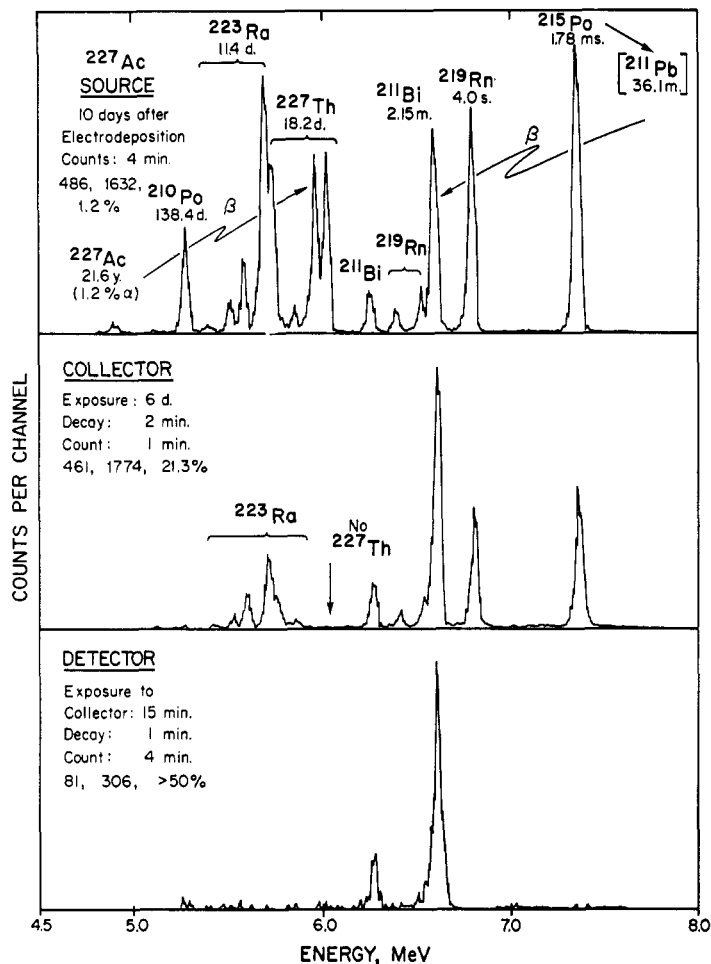
Figure 11. Recoil atoms from aged ²²⁷Ac containing daughters

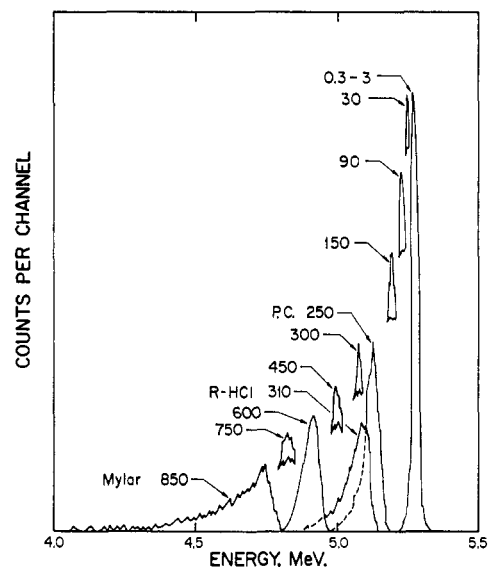Figure 12. Effect of absorber on resolution and apparent energy of ²¹⁰Po

Numbers refer to thickness in $\mu g/cm^2$. P.C., R-HCl, and Mylar refer to commercial films of polycarbonate, rubber hydrochloride, and Mylar, respectively; other numbers refer to air

To decrease possible contamination from polonium present in a sample being analyzed for other alpha-emitters, an attempt was made to volatilize the polonium by heating the stainless steel plates before analysis. However, polonium is surprisingly nonvolatile from stainless steel. One of the aged plates lost only 4.2% of its activity on heating over a blast lamp to a bright red color for 1 minute. Six more successive heatings gave an average loss of only about 1% per minute at red heat. A similar source freshly electrodeposited lost 8% during the first 1-minute heating and 4% in a second 1-minute period. Spontaneous volatilization at room temperature in a vacuum for the latter ignited source was below the detection limit. Another source gave a volatilization rate of 9×10^{-5} cpm per hour exposure per cpm of source activity before ignition and $<5 \times 10^{-8}$ unit after heating just to dull red heat and cooling immediately. The resolution decreased to 28 keV (FWHM) with a 50 mm² detector capable of giving 14-keV resolution. However, ignition at red heat is undesirable because lead, if present, will be volatilized and will carry other nonvolatile components with it, causing serious loss. Fortunately, the spontaneous volatility of even freshly-deposited polonium can be reduced at least to $<5 \times 10^{-8}$ cpm per hour of exposure per cpm of source activity by heating the source plate on an uncovered hot plate for 5 minutes without resultant decrease in resolution or loss of lead or polonium. It seems likely that air oxidation of the deposit to a relatively nonvolatile oxide, occurring slowly at room temperature or rapidly at higher temperature, is responsible for the decrease in volatility observed.
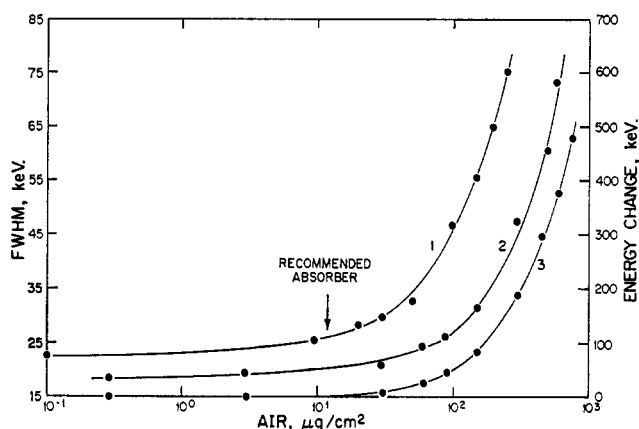
The spontaneous volatility at room temperature in a vacuum of polonium deposited on silver or platinum was less than 2×10^{-7} cpm per hour of exposure per cpm of source activity at 1-mm distance and 0.1-mm pressure, regardless of the time since preparation. It is not known whether the lower volatility is peculiar to the use of noble metals or to the method used to prepare the source. However, the lower volatility of polonium on noble metals is confirmed by the contamination observed from the work with ²¹⁰Po reported below. A source containing 1×10^5 cpm of ²¹⁰Po that had been freshly electrodeposited on polished platinum was used for about 4 hours at distances of ³/₁₆ inch and ⁵/₈ inch from the detector at various pressures between 0.1 and 200 mm of mercury. Contamination of the detector from such extensive use of such a high level source was 50 counts in 1000 minutes, or an average of about 1×10^{-7} cpm per hour exposure per cpm of source activity.

Polonium deposited on platinum using hydroquinone is much more volatile at high temperature than from stainless steel. A plate ignited momentarily just to dull red heat lost 11% of its activity. After 1 minute at bright red heat, the source lost 87% of what remained. After another 1 minute at bright red heat, another 73% of the remaining activity was volatilized. These results indicate little, if any, tendency for the polonium to be converted to a less volatile form.

Because of its relatively long half-life of 138.4 days, any ²¹⁰Po contamination will persist for an untenably long time and special precautions must be taken to avoid its occurrence. Any deliberate spectrometry with ²¹⁰Po sources on silver or

Table I. Effect of Air Pressure on Resolution, Counting Rate, and Apparent Energy of Polonium-210 and Recoil Contamination of the Detector from Actinium-225

Air pressure, mm Hg	Polonium-210				Actinium-225	
	Air thickness, $\mu g/cm^2$	FWHM, keV	Total counts ($\times 10^4$)	Apparent energy, MeV	Air thickness, $\mu g/cm^2$	Recoil activity, c/4 min
0.1	0.3	18	3.25	5.305	0.2	1694
0.5	...	...	...	...	1.0	1573
1.0	3.0	19	3.27	5.305	2.0	1482
2.0	...	...	...	...	4.0	1063
3.0	...	...	...	...	6.0	658
4.0	...	...	...	...	8.0	503
5.0	...	...	...	...	10	90
6.0	...	...	...	...	12	8
7.0	...	...	...	...	14	6
10	30	21	3.25	5.296	20	5
20	60	24	3.29	5.278	...	...
50	150	31	3.25	5.224	100	13
100	300	47	3.26	5.116	200	15
150	450	61	3.24	5.008	...	...
200	600	73	3.25	4.927	...	...
250	750	90	3.23	4.828	...	...

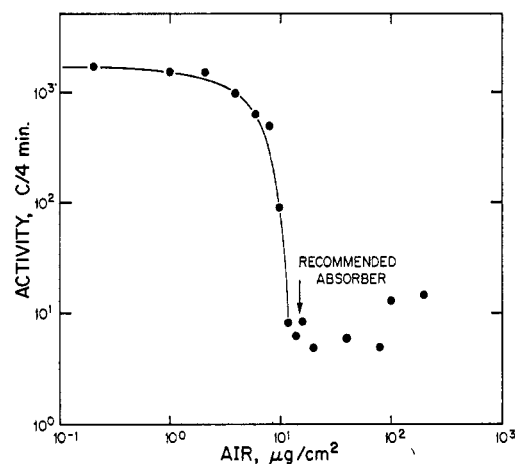
Figure 13. Change in resolution and apparent energy of ^{210}Po with increasing air pressure

Curve 1. Resolution obtained with a source-to-detector distance of 0.64 cm (top shelf) (left ordinate)
Curve 2. Resolution obtained with a source-to-detector distance of 1.92 cm (3rd shelf) (left ordinate)
Curve 3. Shift in peak position or apparent energy. (right ordinate)

Figure 14. Decrease in recoil contamination of ^{221}Fr with increasing air pressure

platinum should be made with as weak sources for as short a time as possible. Stainless steel plates used for the routine identification of alpha-emitters should be heated on an uncovered hot plate for 5 minutes to guard against serious contamination of the detector due to the possible presence of ^{210}Po in the sample. If the contamination were directly proportional to the activity of the source, time of exposure in a vacuum, and number of samples, the maximum rate found above would give 0.003 cpm of contamination from a single 1-hour exposure to a source containing only 20 cpm of ^{210}Po. This level of contamination is just detectable in a 1000-minute spectrum with a clean detector. Because of the essentially additive nature of contamination with 138-day ^{210}Po over a period of several months, very few such samples or a relatively small increase in either exposure time or activity would contaminate the detector to an unacceptable extent.

Prevention of Recoil Contamination. Because of the differences in size and energy, it should be possible to select an absorber sufficiently thin to permit the alpha particles to pass through virtually unaffected while reducing the number of recoil atoms reaching the detector significantly. Most commercial films are so thick that the alpha spectra are degraded untenably, and so flimsy that they are difficult to handle. In contrast, any desired thickness of absorber can be obtained easily and conveniently by merely adjusting the air pressure inside the spectrometer chamber. Figure 12 shows the effect of various thicknesses of air and certain films on the resolution and peak position of a ^{210}Po source using the 50-mm^2 detector. The numerical data obtained from the spectra were used in a computer program to give the resolution (FWHM), the total number of counts under the peak, and the apparent energy of the peak at its high energy edge for the ^{210}Po measurements shown on the left side of Table I. There is little change in either resolution or apparent energy below about 30 $\mu g/cm^2$ of absorber. The total number of counts under the peak remained statistically constant regardless of the shape or position of the peak.

The change in resolution and apparent energy with air pressure is plotted in Figure 13. In addition to the type, surface polish and absorption of the source plate, scattering from the source into the detector and the absorbing medium between the detector and the source also affect the resolution of the detector. Curves 1 and 2 show the poorer resolution obtained on the top shelf compared to the third with equal

absorption because of the greater scattering from the mounting plate and sample into the detector at shorter distances. These curves demonstrate graphically why sources should be placed at least 1.5 diameters from the detector to obtain optimum resolution.

The effect of air pressure on the recoil contamination of ^{221}Fr obtained with the collector plate 1.28 cm from the ^{225}Ac source is shown in Figure 14 and on the right side of Table I. When the thickness of the air path was increased to 12 μg/cm^2, the observed activity was not statistically different from the background of the detector because of contamination from previous runs. Consequently, recoil collection was probably reduced by a factor of at least 10^3 with less than 1 or 2 keV loss in resolution of the alpha spectra. A Cartesian diver can be installed in the vacuum line to obtain and maintain the air pressure automatically at any desired level.

To prove that other source-to-detector distances can be used, similar measurements were made on other shelves. The pressure was adjusted with the source on each shelf until the activity of the recoiling atoms collected became statistically zero in the counting time used. The results are shown in Figure 15. The range of the recoiling atoms was between 12 and 16 μg/cm^2 for all distances checked.

Because all the foregoing recoil collections were made on steel plates in a separate chamber to eliminate contamination of the detector and wall effects in the spectrometer, the adequacy of the final conditions for preventing recoil contamination was tested at the end of the work in the actual chamber used for obtaining alpha spectra. Surprisingly, the procedure failed completely, i.e., nearly as many recoil atoms were collected at 12 μg/cm^2 of air as at 0.3 μg/cm^2. Significantly, recoil activity also reached the detector even when an impenetrable barrier was placed between the source and the detector. Finally, it was found that a negative potential of as little as 2 volts applied to the source plate would permit the two-chamber results to be duplicated in single-chamber use. Application of even several hundred volts had been shown previously to produce no significant decrease in recoil contamination at pressures below 0.1 mm at the short distances employed. Apparently, the air pressure is necessary to absorb the recoil energy and bring the atoms to rest. However, the residual atoms are positively charged and are attacted toward the detector during the counting period because of the charge existing on the detector during use. Even when a barrier is placed between source and detector, the rest ions apparently drift around the barrier and are then attracted to the detector. Only a slight negative potential is required to reverse the situation and attract the positive ions back to the original source plate. A small 6-volt battery can be mounted conveniently inside the sample chamber. When the recoil atoms
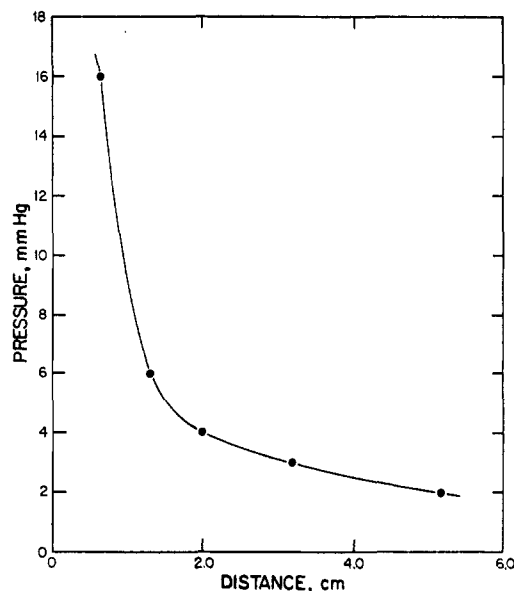

Figure 15. Pressure required to stop recoiling ^{221}Fr atoms at various source-to-detector distances

were collected in a separate chamber, there was no potential on the collector plate as there is on a detector in use, and the rest atoms were flushed out and lost when the collector plate was transferred to the other chamber for counting.

After the present work had been completed, a previous article (9) came to our attention in which similar recommendations were mentioned briefly but few details were given. These authors obtained a reduction in recoil collection of only a factor of 50 at a pressure of 3 cm of mercury, a source-to-detector distance of 1 cm, and 225 volts on the source plate. The resolution was reduced from 17.0 to 20.5 keV with a 25 mm^2 detector. No examples of recoil products were given.

ACKNOWLEDGMENT

(9) A. Chetham-Strode, J. R. Tarrant, and R. J. Silva, IRE Trans. Nucl. Sci., NS-8, No. 1, 59 (1961).