



Delayed Isomeric State in ^{205}Pb and Its Implications for $4\pi\alpha$ Liquid Scintillation Spectrometry of ^{209}Po

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Assays of ^{209}Po by $4\pi\alpha$ liquid scintillation (LS) measurements are confounded by an α -transition to a 2.3 keV ($J^\pi = 1/2^-$) level in ^{205}Pb which was previously unknown to be a delayed isomeric state. The level is believed to be populated by a substantial fraction of the total ^{209}Po decay ($\sim 80\%$), and presumably decays predominantly by internal conversion. The resulting conversion electrons from this delayed isomeric transition were identified in LS spectra. The lifetime is inferred to be *at least* several times 10^{-5} s, although it may very well be substantially longer lived by orders of magnitude. The implications of these findings for routine measurements of ^{209}Po by LS are discussed. Suggested experiments to precisely determine the lifetime of the state are also proposed.

Introduction

Collé *et al.* (1994) recently reported on the preparation and calibration of a new series of carrier-free ^{209}Po solution standards which will be disseminated as National Institute of Standards and Technology (NIST) *Standard Reference Material SRM 4326*. In

the course of that work, primarily as a result of initially inexplicable and confounding difficulties with the $4\pi\alpha$ liquid scintillation (LS) measurements that were used to perform the ^{209}Po calibration, evidence emerged for the existence of a ^{205}Pb delayed isomeric transition. This paper provides further evidence and arguments to support this discovery.

A partial decay scheme for ^{209}Po , based largely on the recent critical evaluation of Martin (1991) and as was currently known at the time of this work, is shown in Fig. 1.† As indicated therein, the decay of ^{209}Po is predominantly ($\sim 99\%$) by two α -particle transitions ($E_{\alpha 0} = 4.885$ MeV and $E_{\alpha 1} = 4.883$ MeV) to the ground state ($J^\pi = 5/2^-$) and first excited state ($J^\pi = 1/2^-$) of ^{205}Pb . The relative α -branching of these two transitions is largely unknown. The two most complete studies of ^{209}Po α -decay by Hagee *et al.* (1966) and Mandal *et al.* (1989) could not resolve the two α emissions, and thereby could only determine a combined transition probability for populating both levels. The assumed relative populations of $\sim 20\%$ to the ground state and $\sim 80\%$ to the 2.3 keV level are based on estimates by Martin (1991) using the systematics of α -decay hindrance factors. Irrespective of the absolute α transition intensity to the 2.3 keV level, it seems clear that it is probably populated to a substantial extent. Furthermore, it is now evident (as detailed below) that the decay of this state is delayed, and not in a prompt coincidence with the α feeding of it. The resulting $\Delta J^\pi = 1/2^- \rightarrow 5/2^-$ transition must be very highly converted with the emission of discrete low energy conversion electrons (*ce*)‡. If the

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†A re-evaluation of the intensities for the principal α and electron capture branches in ^{209}Po decay was recently performed by Schima and Collé (1994) in a companion study.

‡Theoretical calculations of internal conversion coefficients for such a low energy transition are unavailable. One may however appreciate the hindrance to γ -ray emission by considering the so-called Weisskopf "single particle" nuclear model (which assumes such weak coupling between the constituent nucleons that in a γ -transition only a single nucleon (a proton) experiences a change in its quantum state, the specific change assumed in the calculation is $J_{\text{initial}} = l + 1/2$ to $J_{\text{final}} = l/2$). From Weisskopf (1951) and Blatt and Weisskopf (1952), the transition probability for either electric radiation of multipole order $E(\Delta J = l)$ or for magnetic radiation of multipolarity $M(\Delta J = l)$ having an energy E_γ may be expressed in terms of a reciprocal mean lifetime $1/\tau$. The *minimum* mean lifetime of the nuclear state J_{initial} against a γ -transition of multipolarity $E2$ having an energy $E_\gamma = 0.0023$ MeV in $A = 209$ (and with a "statistical factor" $S = 2(1 + 1/l)/[(2l + 1)(2l - 1) \dots (5)(3)(1)](1 + 1/3)^2 \approx 0.005$) is roughly estimated to be $\tau \approx 90$ s (or ~ 60 s on a half-life basis). This independent

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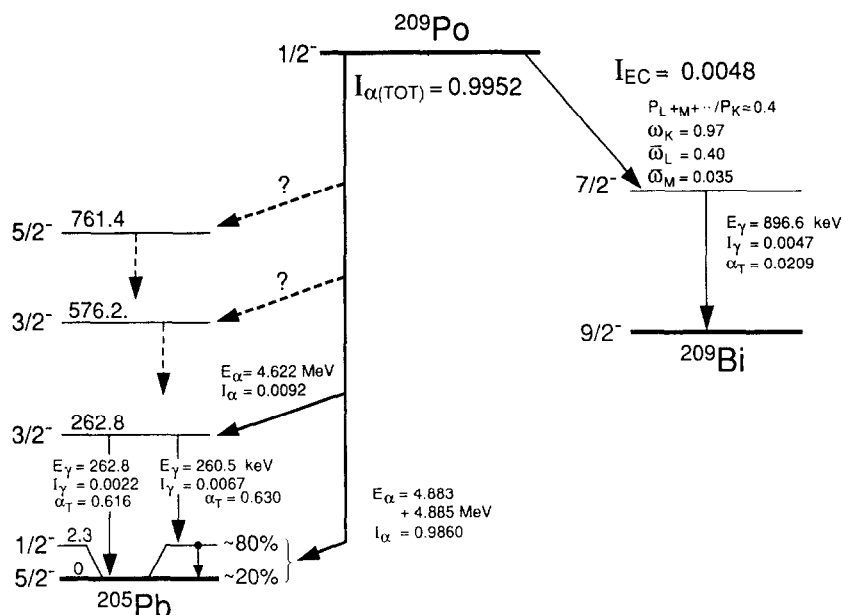


Fig. 1. Partial decay scheme for the ^{209}Po α - and electron capture branch decays.

lifetime of this state is comparable or longer lived than the pulse resolving time of an LS counting system, then some fraction of these *ce* (and attendant 2.3 keV γ -rays, if any) would be detected independent of the feeding α -branch. The measurement situation in this case would be very unlike that for usual $4\pi\alpha$ LS spectrometry of pure (or nearly pure) α -emitting radionuclides, where the LS detection efficiency of α -particles is virtually 100% and independent of reasonable sample quenching conditions. Rather, the apparent combined α and *ce* LS detection efficiency would be greater than 100%, and the efficiency for the *ce* would be dependent on sample quenching and on the specific discrimination levels or detection thresholds for the LS instrument.

Experimental evidence to support the above-described contention that the 2.3 keV isomeric transition in ^{205}Pb is delayed may be summarized as follows:

- (i) A low energy peak roughly centered at around 3 keV (on an equivalent β energy scale) was observed in LS spectra of ^{209}Po . The peak was evident in spectra obtained with two different LS spectrometers and

under wide variations in measurement, sample preparation, and chemical quenching conditions.

- (ii) The large relative magnitude of this ascribed *ce* peak excludes the possibility of it arising from some other known decay mode in ^{209}Po (e.g. from Auger electrons in the electron capture branch to ^{209}Bi).
- (iii) The peak cannot be attributed to a radionuclidic impurity since the ^{209}Po source material, based on high-resolution α - and photon-emission spectrometry, was shown to be very pure.
- (iv) The observed peaks were responses to true events (detected coincident pulses) in the scintillator, and were not due to LS counting system random "noise".
- (v) LS spectra obtained with α - β pulse decay-time discrimination clearly assigned the peaks to β (i.e. electron) pulses.
- (vi) Detailed spectral investigations as a function of widely varying chemical quenching conditions verified that the relative magnitude of this ascribed *ce* peak (reflecting its apparent detection efficiency) was quench dependent. When this attributed *ce* component was subtracted from the total LS response and spectra, one obtained an invariant ^{209}Po α -emission rate that was independent of quenching.
- (vii) Based on comparative measurements of the same samples with two LS spectrometers, the relative magnitude of the ascribed *ce* peak was invariably larger with the spectrometer having a shorter coincident pulse resolving time. One cannot, however, completely ex-

Footnote continued—

particle model estimate is only a crude order-of-magnitude approximation, and does not account for any decay by internal conversion. A correction for internal conversion would, of course, result in a diminution of the decay lifetime. Furthermore, the mean lifetime could very well be one or two orders of magnitude less than this estimate since known E2 transition probabilities (for much higher transition energies) are typically found to be larger than single-particle approximations by factors ranging from over 100 to near unity. The possibility for an MI admixture only blurs the picture further.

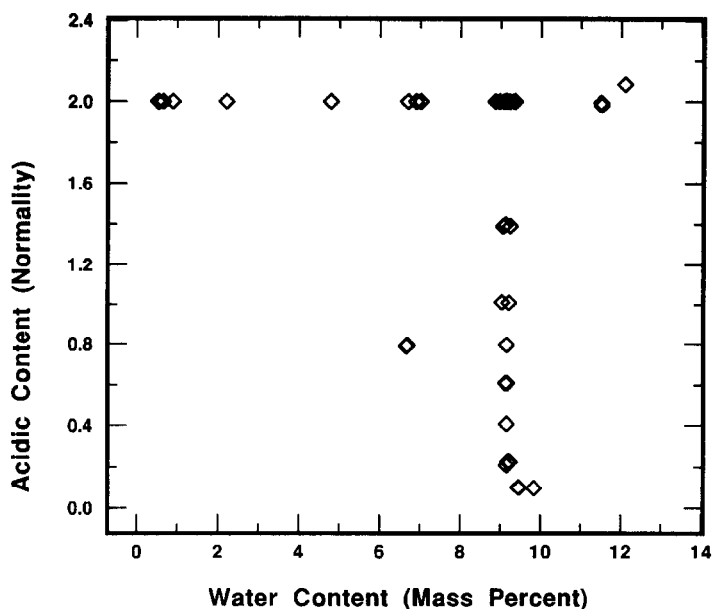


Fig. 2. Compositions of the LS sources investigated for this study. The two composition variables are p_{aq} (abscissa) which is the total aqueous phase content of the cocktail in mass percent, and N_{aq} (ordinate) which is the acidic normality in the aqueous phase. Total source masses ranged from approx. 10–15 g (with a range of 0.1–1.0 g of ^{209}Po sample solution in 10–14 g of scintillation fluid). The relation between the source compositions and corresponding quench indicating parameters are addressed at length in Collé *et al.* (1994).

clude the possibility that this was rather a result of differences in the discrimination thresholds for the two instruments.

- (viii) $4\pi\alpha$ LS measurements of ^{209}Po α -emission rates were in excellent agreement with confirmatory measurements by $2\pi\alpha$ gas-flow proportional counting, *but only after the ascribed ce response was subtracted from the LS spectra.*

The above findings are largely based on studies whose experimental aspects are described in detail in Collé *et al.* (1994). These studies included exhaustive LS measurements and spectral examinations, the α - and photon-emission impurity analyses, and $2\pi\alpha$ confirmatory measurements. The LS results, which are mainly considered here, were obtained over the course of approx. 14 months of investigation; from nearly 500 independent LS measurements on each of two different LS spectrometers; and using over 40 different LS samples, which were in turn obtained from nine different ^{209}Po solutions having gravimetrically-related concentrations.

Experimental Results and Discussions

The LS samples (along with matched blanks of nearly identical composition for background subtrac-

tions) covered a diverse range of sample compositions and chemical quenching conditions, including samples that were unstable cocktail mixes that exhibited large temporal variations in chemical quenching. Figure 2 illustrates a matrix of sample compositions that were studied for this work. As shown, the majority of samples had compositions composed of two cross sections across the two p_{aq} and N_{aq} variables: a nearly constant mass percent of water at p_{aq} of 9–10%, with varying acidic normality N_{aq} ; and $N_{\text{aq}} = 2\text{ N}$, with varying p_{aq} . Refer to Collé *et al.* (1994) for further sample composition details.

The two LS counting systems that were used are here designated at “*Beckman*” and “*Packard*”.* They were also described in considerable detail earlier (Collé *et al.*, 1994). Both employ two high-performance, matched photomultiplier tubes (PMT) operating in a coincidence mode with pulse summing. Their main differences are twofold: in their pulse amplification, and in their quench indicating parameters (QIP). The *Beckman* utilizes a logarithmic pulse amplifier coupled to an analog-to-digital converter (ADC) for spectral pulse-height analyses, while the *Packard* uses linear amplification for its ADC pulse-height analyses. The *Beckman* contains an external ^{137}Cs source for Compton-edge (Horrocks number) quench monitoring. The Horrocks number ($H^\#$) QIP is based on the downward spectrum shift of the Compton edge of the external ^{137}Cs γ -ray source with increasing sample quenching. The parameter corresponds to the spectral channel number

*The mention of commercial products within this paper does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products identified are necessarily the best available for the purpose.

shift between the quenched sample and an unquenched blank sample. The channel number shift ($c_2 - c_1$) is, because of the logarithmic amplification, equal to a logarithmic energy ratio $\log(E_2/E_1)$. The *Packard* has an external low-energy ^{133}Ba γ -ray source that is used to obtain a "transformed Spectral Index of the External Standard ($tSIE$)" quench indicating parameter. This QIP, $tSIE$, is based on a proprietary mathematical transform of the energy distribution of the ^{133}Ba generated Compton spectrum, and consists of a relative quenching scale in which unquenched samples correspond to $tSIE = 1000$. Correlations between the QIPs for the two spectrometers, as applied to the samples considered here, were addressed by Collé *et al.* (1994). Resolving times for coincident pulses in the two spectrometers were also somewhat different. The *Packard* had a fixed (i.e. "non-extending") resolving deadtime of $\tau \approx 12 \mu\text{s}$, whereas the *Beckman*'s was variable (dependent on pulse height) from about 5–33 μs . The latter spectrometer would of course spend the vast majority of its time counting the principal ^{209}Po α . Using the pulse heights for the dominant α -peak (i.e. its channel number locations), the typical resolving times for the *Beckman* were $\tau = 15 \pm 4 \mu\text{s}$ for most of the spectra considered here.

A comparison of typical ^{209}Po spectra obtained with the two spectrometers is shown in Fig. 3. The spectra were appropriately normalized to the same abscissa and ordinate scales. The conversions of spectral channel numbers to a beta energy scale are, at best, only very approximate since the channel

number to energy conversions are dependent on sample quenching (Collé *et al.*, 1994). Furthermore, it is difficult to directly compare the spectra since the *Beckman* instrument with its logarithmic amplification gives ADC channel numbers corresponding to a logarithmic energy scale while the conversion of channel number to energy is nominally linear with the *Packard*. This then requires normalizations of both the energy axis as well as the count rate *per channel* (or *per keV*). The energy intervals per channel are approximately of equal width in the *Packard*, but are of unequal logarithmic widths in the *Beckman*. Both spectra shown in Fig. 3 were obtained with the same sample and had respective QIP values of $H^* = 73$ and $tSIE = 528$. The sample consisted of about 0.13 g of a ^{209}Po master solution (nominal 2 N HCl) in 14.1 g of scintillator, corresponding to $p_{\text{aq}} = 0.89\%$ and $N_{\text{aq}} = 2 \text{ N}$ (see Fig. 2 for comparisons). The LS response from the dominant 4.88-MeV α of ^{209}Po appears as a peak at around roughly 450–550 keV in the corresponding β energy scale. The peaks centered at approx. 3 keV, as shown on the expanded scale inset, is attributed to the ^{205}Pb conversion electrons. One may observe that in the region below 10 keV, the *Packard* spectrum has only nine or ten channels of data while that from the *Beckman* instrument has over 50. Other details of the spectra are considered at length elsewhere (Collé *et al.*, 1994). The ratios of the integral area of the electron peak to that for the α -peak, R_e/R_α , are 0.059 in the *Packard* and 0.029 in the *Beckman*. At the same time, values of R_α for the two spectra are in agreement to better than 0.2%. The large difference in the two R_e/R_α values may be

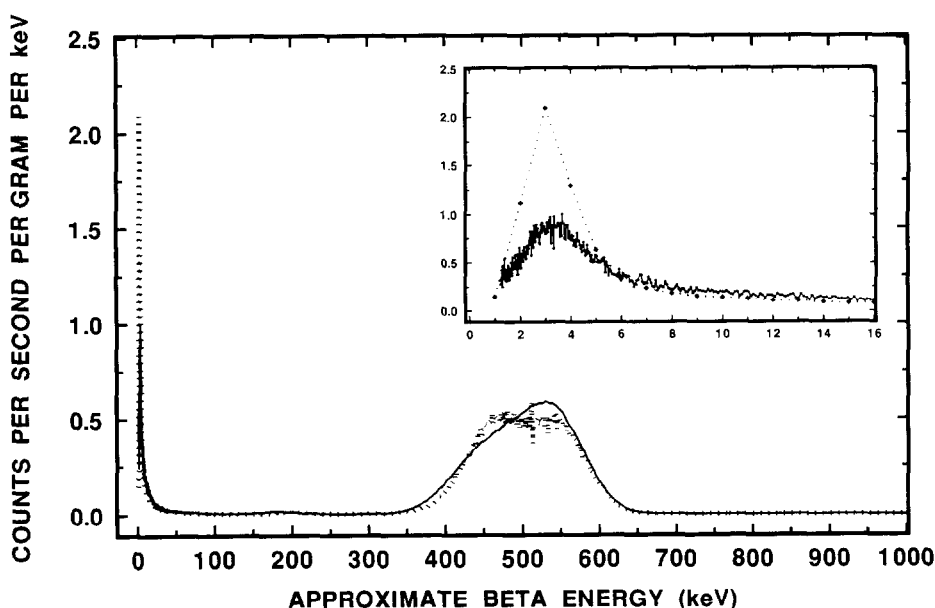


Fig. 3. Comparison of typical ^{209}Po LS spectra obtained with the *Beckman* (—) and *Packard* (· · ·) systems. The broad peaks at around an equivalent β energy of 500 keV are the responses from the ^{209}Po alphas. The inset on an expanded energy scale shows the peaks that are ascribed to conversion electrons from the delayed 2.3-keV isomeric transition in ^{205}Pb .

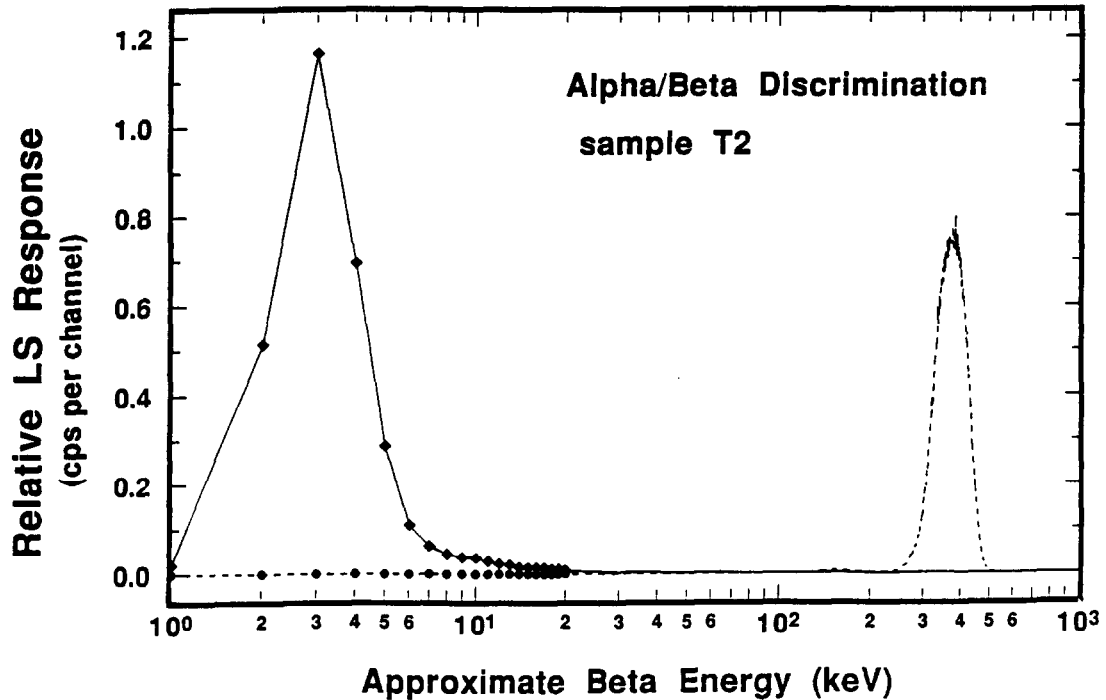


Fig. 4. Typical ^{209}Po spectra obtained using α - β pulse decay discrimination. The β (electron) spectrum showing the low energy peak that is attributed to ce from the 2.3-keV isomeric transition in ^{205}Pb is given by the solid line, and the α spectrum by the broken line. The relative magnitude of the integral areas of the two peaks is $R_c/R_\alpha = 0.042$.

ascribed to differences in detection and resolution of the ce by the two instruments. The larger ratio was, as in all other spectral comparisons, obtained with the *Packard* that had a shorter resolving time. The differences, however, could equally be due to different discrimination thresholds in the two instruments (Houtermans, 1973).

Based on α - β pulse decay discrimination the low energy peak noted above could definitely be attributed to true coincidences of β (electron) selected pulses. Typical results of the composite α spectrum (---) and β spectrum (—) are given in Fig. 4. As shown, the low energy peak (again centered at around 3 keV) is clearly due to β -pulses and is considered to be the result of conversion electrons of discrete energies.* In contradistinction, only α selected pulses comprise the ascribed broad α -peak. The relative magnitudes of the conversion electron (R_c) and α (R_α) peaks shown in the spectra may be deceiving on initial impressions. One should note that the ordinate is in units of a relative counting rate per channel (which corresponds in this case to a counting rate per

keV), and that the abscissa is a logarithmic energy scale. The R_c/R_α ratio in the spectra of Fig. 4 is in fact 0.058. Further experimental details for these α - β discrimination experiments may again be found in Collé *et al.* (1994).

The detection efficiency for this attributed ce peak was distinctly dependent on sample composition and chemical quenching. Figure 5 provides the ratio R_c/R_α as a function of N_{aq} for one series of samples (with nearly constant $p_{aq} \approx 9.1\%$) as obtained with both LS spectrometers. The variable N_{aq} was used as a measure of chemical quenching and was highly correlated with the QIP values for the instruments as shown in Fig. 6. The relative magnitude of R_c (as a percentage of R_α), given in Fig. 5, ranges from about 4 to 2% with the *Packard*, and from about 1.6 to 0.8% with the *Beckman*. Again, larger R_c/R_α values were invariably obtained with the *Packard* instrument. In addition, the rate of change in R_c as a function of quenching was invariably greater with the *Packard* data. The latter finding would tend to support that the R_c/R_α differences between the two instruments were more likely a result of detection threshold differences than due to resolution time differences. In this same series of samples, the R_α -values obtained with either instrument were invariant. Figure 7 clearly depicts the invariance of the α -detection efficiency with chemical quenching and between instruments. In Figs 5–7, the data are based on five replicate measurements of each sample with

*Only ejection of N, O and P shell electrons is energetically allowable. The low 2.3 keV transition energy precludes ejection of all K, L and M shell electrons (with binding energies ranging from $B_K = 88.0$ keV to $B_{MV} = 2.48$ keV). Kinetic energies of the ce can range from a minimum of roughly 1.4 keV (for the N_1 subshell electrons) to a maximum of about 2.3 keV (for the P subshells).

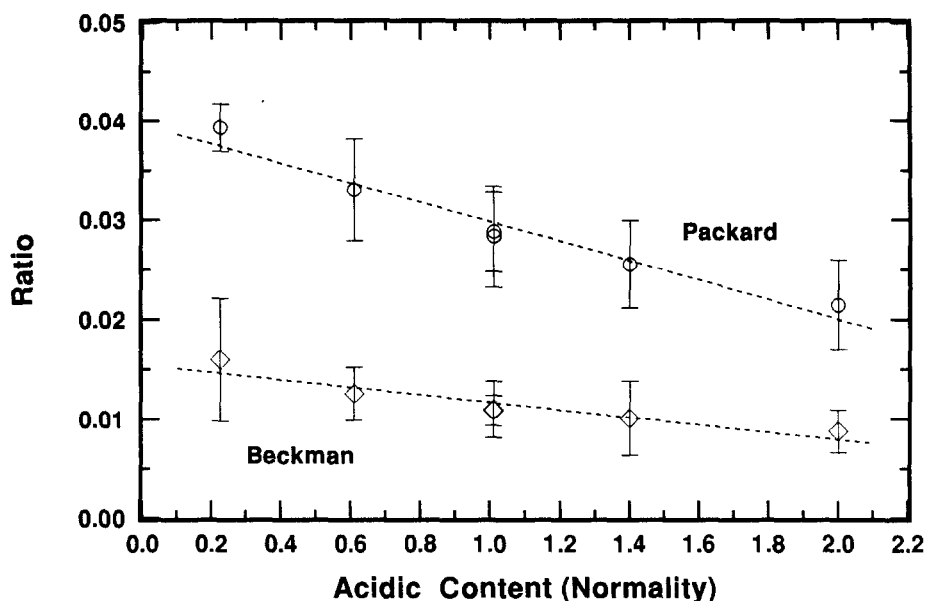


Fig. 5. The ratio of R_e/R_s as a function of variable N_{aq} in a series of samples as obtained with the *Packard* and *Beckman* LS spectrometers.

the *Beckman* instrument, and 10 measurements on each sample with the *Packard*. The error bars on the individual mean datum points in Figs 5 and 7 correspond to calculated standard deviations for the five or ten replicate measurements. The mean R_s across all samples and with both instruments, $R_s = 85.48$ counts $s^{-1} g^{-1}$, is also shown in Fig. 7 as a broken line. The dotted lines correspond to upper and lower limits for a ± 1 SD uncertainty interval about this mean. The apparent poorer measurement precision in R_s for the *Beckman* data is merely a result of fewer

measurements (degrees of freedom) for each sample with the *Beckman*. Comparable results for R_e and R_s as a function of variable quenching (using other sample composition variables) were obtained for all of the other sample series previously considered (Collé *et al.*, 1994). For example, an M series of 12 samples (three samples from each of four identical ^{209}Po standard solutions), all having nearly constant $p_{aq} \approx 9.1\%$ and $N_{aq} = 2$ N, based on 10 replicate measurements of each sample on each instrument, gave grand means R_s counting rate concentrations

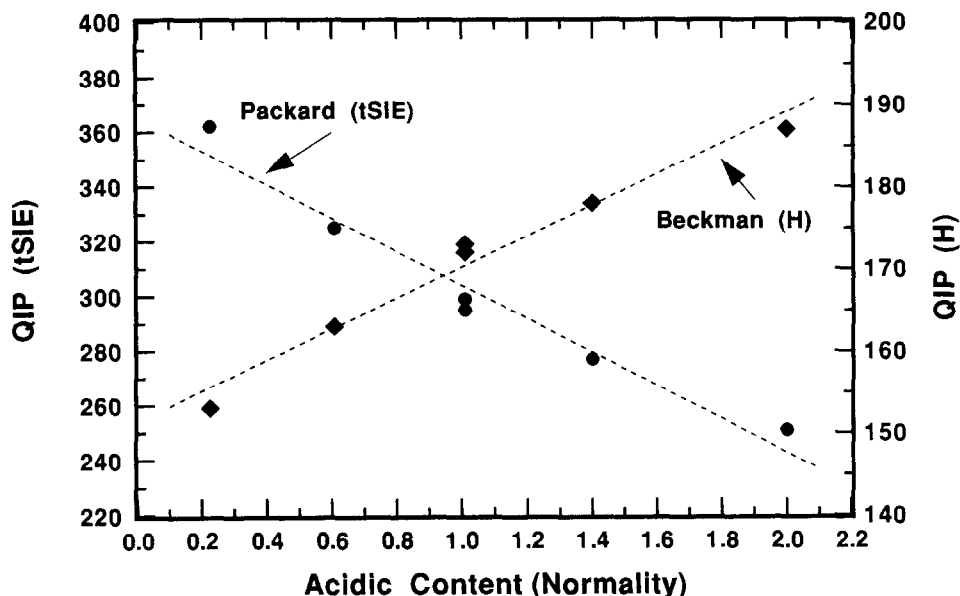


Fig. 6. Relationships between the quench indicating parameters QIP (H^* for the *Beckman* instrument and $tSIE$ for the *Packard*) and N_{aq} for the sample series of Fig. 5.

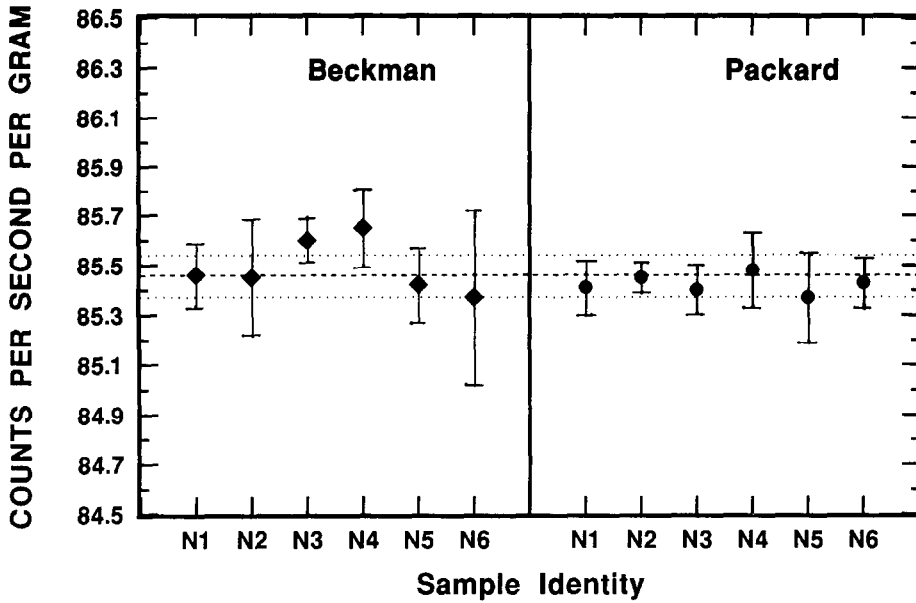


Fig. 7. The mean α counting rate concentration R_α obtained with both LS spectrometers for the sample series of Figs 5 and 6. The invariance in the α -detection efficiency between samples (with varying quenching) and between instruments is evident.

of $R_\alpha(\text{Beckman}) = 85.43 \pm 0.25 \text{ counts s}^{-1} \text{ g}^{-1}$ and $R_\alpha(\text{Packard}) = 85.43 \pm 0.23 \text{ counts s}^{-1} \text{ g}^{-1}$ with R_e/R_α ratios of 0.017 and 0.027, respectively. The above uncertainty intervals for the R_α means are standard deviations of the means with 119 df. An extreme case of the effect of chemical quenching is illustrated in Fig. 8 for an unstable LS sample that

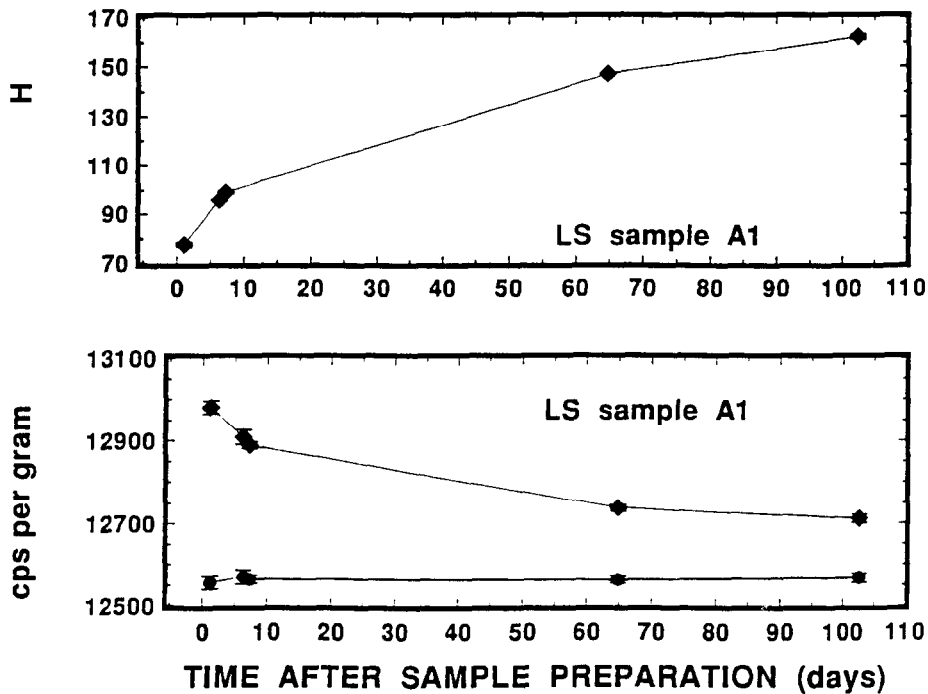


Fig. 8. Quench parameter H^* (upper trace) and LS counting rate concentrations $R_{\alpha+\epsilon}$ and R_α (lower trace) for an unstable LS cocktail sample of ^{209}Po as a function of the time after sample preparation as obtained from sequential measurements with the Beckman LS spectrometer. The upper curve in the lower trace corresponds to the counting rate concentrations $R_{\alpha+\epsilon}$ obtained from integrating the full-energy LS spectra; the lower curve is for R_α and is that obtained after subtraction of the component LS responses from the 2.3-keV ^{205}Pb isomeric transition. Differences between $R_{\alpha+\epsilon}$ and R_α correspond to R_e values.

exhibited large temporal variations in quenching. The cause of this sample instability was subsequently believed to be a result of deleterious interactions of traces of unexpelled nitric acid in the ^{209}Po solution with the LS scintillation fluid (Collé *et al.*, 1994). The measurement results for the sample as a function of measurement time had large and not very systematic variations in QIP values and in values of R_c (obtained from the differences in the two $R_{\alpha+ce}$ and R_α curves in the lower trace of Fig. 8). Each plotted value in Fig. 8 is a mean derived from five to ten measurements with the *Beckman* spectrometer. Over a period of approx. 100 days, the quench parameter H^* varied from about 75 to over 160, with the largest changes occurring in the first 10 or so days. Corresponding changes in the *Packard* QIP, $tSIE$, for the same sample over a shorter 55 day period ranged from 386 to 251. Over the course of the measurements with the *Beckman*, the R_c/R_α ratios monotonically decreased from 0.034 to 0.011 while the mean R_α values over the same time interval were constant and independent of the sample quenching changes (i.e. they were statistically equivalent within their measurement precision). In all cases considered (Collé *et al.*, 1994), R_c was found to vary with time, with sample quenching conditions, and with the instrument used to perform the measurement; whereas the R_α -values were always invariant.

The relative magnitudes of the R_c values found in these experiments were quite substantial ranging from just less than 1% to upwards of nearly 6% (depending on the sample and quenching conditions). These magnitudes preclude several possibilities as to the origin of the low energy peaks in the LS spectra. Based on the known relative transition intensities for the EC branch and lower-energy α -branches (Martin, 1991; Schima and Collé, 1994), no alternative ^{209}Po decay mode has a relative intensity that could possibly approach several percent (see Fig. 1). Similarly, based on extensive α - and photon-emission impurity analyses (Collé *et al.*, 1994), detection limits for any possible radionuclidic impurity excluded anything that could conceivably approach magnitudes of several percent. The only observed impurity was a trace of ^{208}Po , having a ^{208}Po to ^{209}Po impurity ratio of 0.00124 ± 0.00010 . Lastly, two wholly independent ^{209}Po calibrations by $2\pi\alpha$ gas-flow proportional counting and by $4\pi\alpha$ LS spectrometry resulted in mean ^{209}Po α -emission rate concentrations of $85.5 \pm 2.6 \alpha \text{ s}^{-1} \text{ g}^{-1}$ and $85.42 \pm 0.15 \alpha \text{ s}^{-1} \text{ g}^{-1}$, respectively. The former was obviously less precise and merely served as an independent measurement confirmation. Nevertheless, had the R_c components been included in the LS results, the two determinations would have been in statistical disagreement. The $4\pi\alpha$ LS determination would have been outside the uncertainty bounds of the $2\pi\alpha$ confirmation. Clearly, the R_c component was detected in addition to and independent of the detection of the R_α component.

All of the above findings are consistent with the

previous assertions that very low energy conversion electrons of considerable magnitude were detected in the decay of ^{209}Po , and that the ^{205}Pb 2.3-keV isomeric state (fed by the α decay of ^{209}Po) is sufficiently long-lived for its conversion electrons to be detected independently of the ^{209}Po α -transitions. No other interpretation of the findings can be deduced.

It is impossible to even crudely approximate the lifetime of the ^{205}Pb state, with the available data, except to establish a lower limit that could very well be underestimated by orders of magnitude. The estimated resolving times τ for the two LS spectrometers were of comparable magnitude, in the general range of 10–20 μs . Therefore, the lifetime of the state must be at least of this magnitude, and probably (for the ce to be reasonably detected) at least several times this magnitude (i.e. at least several times 10^{-5} s). The slight differences in τ cannot be relied upon to account for the observed differences in R_c obtained with the two instruments. Furthermore, the observed differences in R_c are more likely to be a result of differences in the respective instrument's absolute detection thresholds at these low energies, which are largely unknown. Considering the likely detection efficiency for these low energy 1.4–2.3 keV ce (Houtermans, 1973), and that their abundance in terms of the probability per decay of ^{209}Po (assuming that $\sim 80\%$ of the α -decay feeds the 2.3-keV level) is large, the observed R_c values are of such magnitude as to suggest that the lifetime is indeed substantial—manyfold τ .

The implications of these findings for LS measurements of ^{209}Po are easily seen. The apparent LS detection efficiency for ^{209}Po (in terms of counts α^{-1} or counts s^{-1} per Bq of ^{209}Po) will be greater than 100% if one integrates the full-energy LS spectrum. The exact magnitude above 100% will depend on the fraction of detected ce from the ^{205}Pb transition. This fraction will, in turn, be highly dependent on the LS sample composition, chemical quenching conditions, and the characteristics of the LS spectrometer used to perform the measurement. Errors may ensue even if ^{209}Po standards are used as a reference basis for comparative or relative measurements with matched LS samples. As indicated by the variations in R_c or R_c/R_α values given above, the variability in the detected fraction of the ^{205}Pb ce is very sensitive to quench changes. It may be difficult to match the compositions of standards and samples close enough to achieve the desired level of accuracy. One should therefore either exclude or account for the ^{205}Pb ce response when assaying or calibrating ^{209}Po by LS spectrometry. This is not difficult to achieve considering the tremendous separation between the ^{209}Po α and ^{205}Pb ce LS pulse heights. We suspect that it is probably possible to detect the ^{205}Pb ce peak in LS spectra obtained with reasonably quenched samples and most commercial LS spectrometers. One then merely needs to either set a suitable discrimination level of “counting window” above the ce peak prior

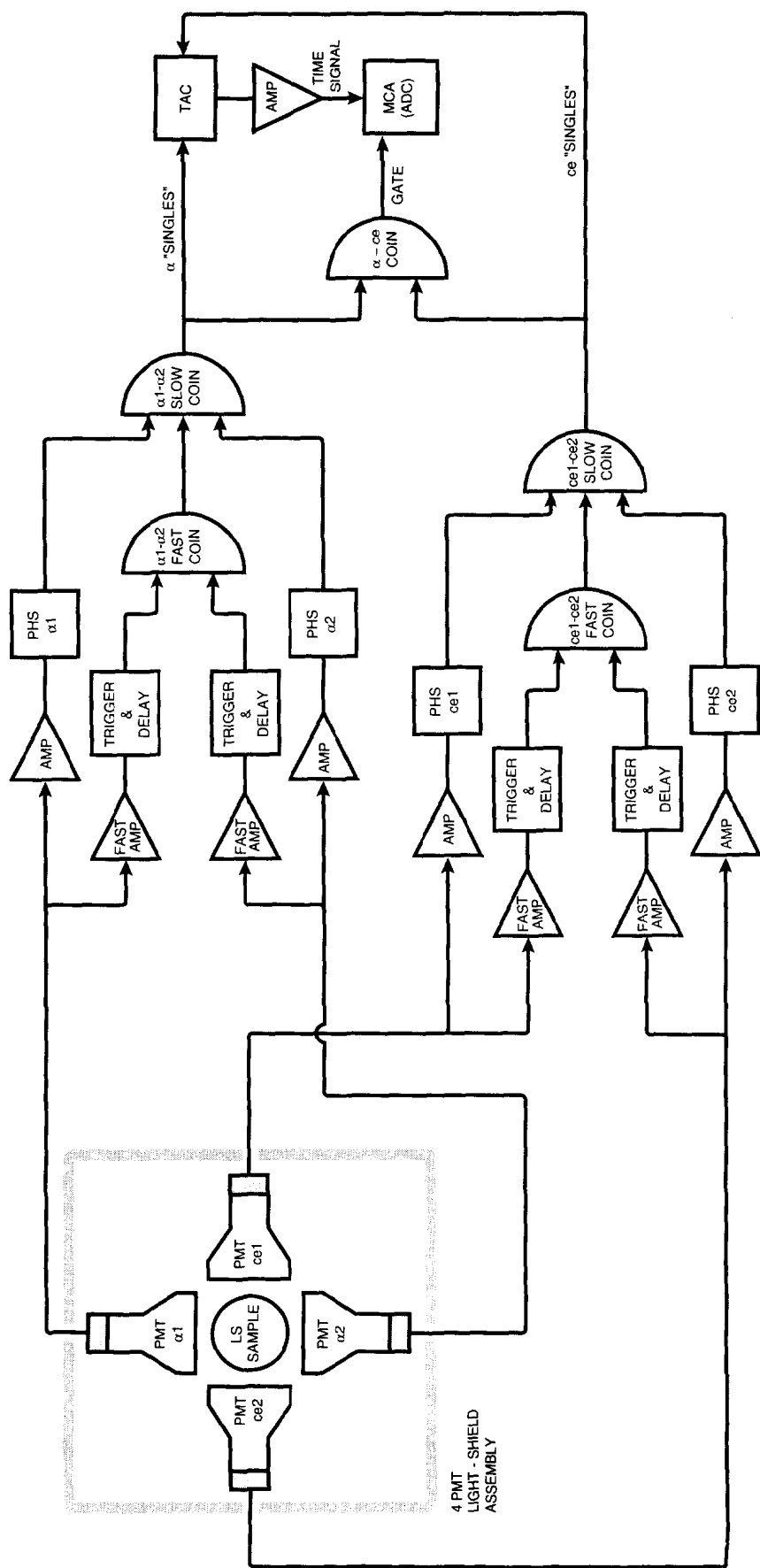


Fig. 9. Schematic of an experimental configuration for a conceivable and proposed determination of the lifetime of the 2.3-keV ($J^\pi = 1/2^-$) level in ^{205}Pb by α - e^+ delayed coincidence measurements. The determination relies upon detection of both the α and e^+ transitions by liquid scintillation, and a conventional multichannel delayed coincidence method using a time-to-amplitude converter (TAC). To minimize high LS background counting rates and improve signal resolutions, the α and e^+ "singles" are obtained from previous fast-slow coincidences of the individual α and e^+ pulses using two sets of paired photomultiplier tubes (PMT). The third coincidence requirement is the α - e^+ coincidence of the α and e^+ coincident pulse "singles".

to counting, or to extrapolate the ^{209}Po α -response under the peak and thereby subtract the ^{205}Pb contribution from the spectrum (Collé *et al.*, 1994). α - β pulse shape discrimination could also be adequately employed. In any case, it would be advantageous to evaluate the specific sample/measurement situation and conditions by first performing careful spectral examinations.

A Proposed Lifetime Experiment

In the course of this work, it became apparent that the lifetime of the delayed ^{205}Pb state could be precisely determined by conventional delayed coincidence techniques (Bell, 1965). Detection of both the ^{209}Po α -emissions and ce from the delayed state by liquid scintillation offers a unique opportunity and straightforward approach to do so. Figures 3 and 4 depict the possibilities of detecting both emissions by LS. The principle advantages of using LS detection are that the detection efficiencies are large, and that the LS counting source preparation requirements are minimal. With conventional solid state (e.g. with surface-barrier junction) detectors, the counting source requirements (and their attendant source preparations) for detection of highly absorbed α -particles and ~ 2 keV ce can be formidable.

In the simplest case, one can easily conceive of using two photomultiplier tubes (PMT), as exists in most commercial LS spectrometers, for the delayed coincidence measurements. One PMT could be used to pulse-height select (PHS) the α -pulses, and the other PMT to PHS the ce pulses. The two pulse-height selected singles for the delayed α -pulses (using an appropriately imposed delay) and for the ce pulses could then be used to form α - ce coincidences. The lifetime is then directly derivable from the coincidence counting rate as a function of the imposed delay time. This essentially is the basic principle and rudiments of a differential delayed coincidence method (Bell, 1965). As a practical matter, with modern modular nuclear electronics, one would be more likely to employ coupled fast-slow coincidence elements*, a multichannel pulse-height analyzer (MCA), and possibly a time-to-amplitude converter (TAC) for the circuitry.

Random electronic "noise" and background LS counting rates from a single PMT are very high. Considerable improvements in the resolution of the α and ce pulses would therefore result from the use of two sets of paired PMT with each pair operating in coincidence. Figure 9 illustrates a proposed experimental configuration for such a four-PMT arrange-

ment that could be used to perform the delayed coincidence measurement. It is based on and utilizes fast-slow coincidences of pulses from the two sets of PMT to form the α and ce "singles", an α - ce coincidence element to gate a MCA, and a TAC to measure the time interval between the two "singles". For simplicity, the scheme of Fig. 9 does not contain provisions to preserve the spectra (pulse height distributions) of the α and ce "singles". This would be a very desirably feature, and relatively easy to incorporate. The figure also does not display other elements that may be required to fulfill the pulse shaping and timing requirements for the coincidence circuitry. Many alternative schemes could equally be used. For example, a variable linear delay of the α "singles" (imposed before the α - ce coincidence element) could be used instead of the TAC. Alternatively, the $\alpha 1$ - $\alpha 2$ and $ce 1$ - $ce 2$ slow coincidence elements could be replaced by pulse summing of the signals from the two PMT, as is accomplished in most commercial LS spectrometers. The summed ($\alpha 1 + \alpha 2$) and ($ce 1 + ce 2$) pulses would then be coincidence gated by the $\alpha 1$ - $\alpha 2$ and $ce 1$ - $ce 2$ fast coincidence elements prior to the α - ce coincidence. Other schemes for determining the lifetime by LS detection using four PMT are also conceivable.

Conclusions

To conclude, three points may be noted. Firstly, ^{209}Po decays to a substantial extent through a very low energy, delayed isomeric state in ^{205}Pb . We contend that the evidence for this is compelling. Secondly, the presence of the resulting delayed isomeric transition in ^{205}Pb has important repercussions for routine $4\pi\alpha$ LS assays of ^{209}Po . Adequate precautions must be made to either exclude or adequately account for the ^{205}Pb transition's contributions (primarily conversion electrons) to the ^{209}Po LS spectra or counting data. Thirdly, and lastly, a precise determination of the lifetime of the delayed ^{205}Pb state by a delayed coincidence measurement using LS detection with four photomultipliers appears feasible. It is believed to be an intriguing possibility since no delayed-coincidence lifetime determination by a LS spectrometry technique is known.

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*Fast-slow coincidences are based on first performing the coincidence function (with a very fast nanosecond-range coincidence element) on pulses unselected as to pulse height, then performing the pulse-height selection on pulses unselected as to time, and following these with a slow coincidence element (in the microsecond range) to select events that satisfy both criteria.

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