Lightest Isotope of Bh Produced Via the ^{209}Bi(^{52}Cr,*n*)^{260}Bh Reaction

S. L. Nelson,[1,2] K. E. Gregorich,[1] I. Dragojević,[1,2] M. A. Garcia,[1,2] J. M. Gates,[1,2]
R. Sudowe,[1§] H. Nitsche[1,2]

[1] Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720

[2] Department of Chemistry, University of California, Berkeley, California 94720

ABSTRACT

The lightest isotope of Bh known was produced in the new ^{209}Bi(^{52}Cr,*n*)^{260}Bh reaction at the Lawrence Berkeley National Laboratory's 88-Inch Cyclotron. Positive identification was made by observation of eight correlated alpha particle decay chains in the focal plane detector of the Berkeley Gas-Filled Separator. ^{260}Bh decays with a 35^{+19}_{-9} ms half-life by alpha particle emission mainly by a group at 10.16 MeV. The measured cross section of 59^{+29}_{-20} pb is approximately a factor of four larger than compared to recent model predictions. The influence of the N = 152 and Z = 108 shells on alpha decay properties are discussed.

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[§] Present address: University of Nevada Las Vegas, Dept. of Health Physics, 4505 Maryland Parkway, Campus Box 453037, Las Vegas, NV 89154-3037

The synthesis and identification of new transactinide (TAN) isotopes and measurements of their decay properties are essential to an understanding of nuclear stability as well as the prospects of producing higher-Z elements and more isotopes in this region of the chart of nuclides. By investigating these isotopes, we will also gain a better understanding of nuclear masses and are able to determine the location and strength of both spherical and deformed shells in this region. Due to the inherent difficulty in producing TAN isotopes, few data about them are available compared to lighter radioactive isotopes. Theoretical predictions can be tested by comparison with experimental data and used to improve models.

For many years, "cold fusion" reactions utilizing various medium-mass projectiles on ^{208}Pb and ^{209}Bi targets have been instrumental in the discovery of many TAN elements and the discovery of their various isotopes. Shell-stabilized Pb and Bi targets lead to favorable Q-values for compound nucleus formation and relatively low excitation energies (approximately 10 – 15 MeV)[1] when using near Coulomb-barrier bombarding energies. These weakly excited (cold) compound nuclei then de-excite through the emission of only one neutron. Elements 107 – 112[2-5] were discovered via this type of reaction which was also used to synthesize the yet to be confirmed element 113[6]. The ^{209}Bi(^{54}Cr,n)^{262}Bh reaction was used in the 1981 discovery of element 107[7, 8] (bohrium, Bh) at the Gesellschaft für Schwerionenforschung (GSI) in Darmstadt, Germany, and we have chosen to use the new but very similar reaction with a projectile that is only two neutrons lighter in a search for a more neutron-deficient isotope of Bh.

Since the original discovery of bohrium as ^{262}Bh^{g,m} and ^{261}Bh at the GSI, four other isotopes of bohrium have been reported[9-11]. Our discovery of ^{260}Bh extends the observed isotopes from $^{260\text{-}262}$Bh and $^{264\text{-}267}$Bh, for a total of seven known isotopes. A previous report of a 2.6 second spontaneous fission (SF) activity that may be a daughter of ^{260}Bh was published in a conference proceeding[12] but is not reported in the peer-reviewed literature. In addition, the methods utilized in the Oganessian work do not provide sufficient evidence for identification of Z or A.

Masses from the 2003 Atomic Mass Evaluation by Audi, Wapstra, and Thibault[13] were used to estimate the Q-values for various decay modes. ^{260}Bh should decay primarily by alpha emission and possibly by SF and electron capture (EC). The predicted alpha particle decay energy is 10.46 MeV, resulting in an expected alpha particle energy of 10.30 MeV and an unhindered half-life of 490μs[14]. The known decay properties of the subsequent ^{256}Db[15], ^{256}Rf[16, 17], ^{252}Lr[15], ^{248}Md[18], and ^{248}Fm[19] daughter products are illustrated in Figure 1.

In this experiment, the new isotope ^{260}Bh was successfully produced via the new "cold fusion" reaction ^{209}Bi(^{52}Cr,*n*). The Lawrence Berkeley National Laboratory's 88-Inch Cyclotron accelerated a 257.0 MeV beam of ^{52}Cr^{12+} with an average intensity of 0.4 pμA. The beam first passed through a 45 μg/cm^2 ^{nat}C foil used to separate the vacuum of the beamline from the Berkeley Gas-Filled Separator (BGS) He gas; the beam then impinged upon a rotating target wheel containing nine arc-shaped ^{209}Bi targets. The targets consisted of a layer of Bi metal with an average thickness of 441 μg/cm^2 deposited on a 35 μg/cm^2 ^{nat}C backing, and were obtained from the target fabrication lab at the GSI. A thin layer (<9 μg/cm^2) of ^{nat}C was also applied to the downstream side of the target surface to improve infrared cooling. Energy loss of the ions through the system was calculated using the program SRIM2003[20].

The beam energy was chosen based on the optimum energy rule by Świątecki, Siwek-Wilczyńska, and Wilczyński[21, 22]. Using tabulated mass defects from Audi *et al.*[13] and an additional experimentally determined offset for odd-Z compound nuclei[23], the center-of-mass beam energy in the center of the target was calculated to be 202.4 MeV, corresponding to a compound nucleus excitation energy of 15.0 MeV, below the threshold for production of ^{259}Bh via the 2*n* evaporation channel.

The reaction products recoiled out of the thin targets with the momentum of the beam and into the 67 Pa He of the BGS. The BGS has been described elsewhere [24-26]. The average evaporation residue (EVR) charge state was calculated to be 7.8[27]. The BGS magnet currents were chosen to direct the ^{260}Bh recoils with a magnetic rigidity of 2.15 T·m[27] to the focal plane Si-strip detector. Monte Carlo simulations of EVR trajectories in the BGS, as in[27], indicate a total separator efficiency of 65 ± 6%. At the focal plane, the ^{260}Bh EVRs passed through a multi-wire proportional counter (MWPC) filled with isobutane gas before entering the Si strip detector array. The MWPC allowed discrimination of implantation events from alpha-like decay events.

Calibration of the Si strip detector was performed with a retractable four-point alpha source containing ^{148}Gd, ^{239}Pu, ^{241}Am, and ^{244}Cm. The alpha particle energy resolution for implanted nuclei was 55-keV FWHM, determined from a ^{173}Yb(^{30}Si,6*n*)^{197}Po reaction run two weeks prior to this experiment. The vertical position resolution was determined by the method described in earlier experiments[28].

During the irradiations, the rate of "EVR-like events" ($8.0 < E(\mathrm{MeV}) < 24.0$, coincident with MWPC signals and anticoincident with upstream or punchthrough detectors) was 0.3 Hz. The rate of "α-decay like events" ($8.0 < E(\mathrm{MeV}) < 11.0$, focal plane only or reconstructed from focal plane + upstream detector, anticoincident with punchthrough detector and MWPC) was $4.9 \cdot 10^{-3}$ Hz. ^{260}Bh was identified by detection of time and position correlated event chains corresponding to EVR implantation followed by the α-decay of ^{260}Bh and ^{256}Db (and possibly ^{252}Lr), or α-decay of ^{260}Bh followed by the SF of ^{256}Rf, the EC daughter of ^{256}Db. To minimize the contribution of random correlation of unrelated events, a fast beam-shutoff scheme was employed. Upon detection of an EVR-like event followed by a position- and time-correlated (within 3σ and 10 s, respectively) ^{260}Bh-α-decay-like event, the beam was switched off for 180 s to allow a background-free search for any daughter-like decays. (See Figure 1).

Table 1 contains the observed eight decay chains attributed to the decay of ^{260}Bh. Some focal plane events had below-threshold energies from either the top or bottom of the strip. If these "single-ended" events are part of a decay chain, the missing energy from the below-threshold signal can be calculated from the signal from the above-threshold end of the strip by assuming the vertical position is the same as other members of the event chain. These calculated missing energies are denoted by parentheses within square brackets in Table 1, and their positions are marked as either greater or less than 0.0 mm.

Full energy alpha decays were recorded for seven of the eight ^{260}Bh alphas. The remaining ^{260}Bh decay in chain number 4 was an "escape," registering only 770 keV in the focal plane. In addition, a ninth chain was observed as an implantation followed by two escapes, an alpha decay of 9.04 MeV, and another escape. This chain could be attributed to the decay of ^{260}Bh but is not included in these results because of its uncertain nature. Half-life and cross section errors were treated as a special case of the Poisson distribution as in [29]. Using the eight alpha decay lifetimes, the half-life of ^{260}Bh was found to be 35^{+19}_{-9} ms. No direct spontaneous fissions or SF resulting from the EC decay of ^{260}Bh to ^{260}Sg were observed, and we assign an upper limit of <18% for the sum of SF and EC branches. The corresponding partial half-life for SF and EC decay is >192 ms.

There is evidence of a grouping of four alphas (from chains 2, 3, 6, and 7 in Table 1) between 10.13-10.19 MeV, with a mean alpha particle energy of 10.16 MeV. There is also one event each at 10.24, 10.08, and 10.03 MeV. Many alpha particle energies are feasible due to possible population of different states in the odd-odd ^{256}Db daughter. The corresponding alpha decay hindrance factor for the 10.16 MeV group based on the four decays comprising that group is approximately 53[14].

The observed ^{256}Db data are in good agreement with decay data previously reported[15]. Of the eight events corresponding to ^{256}Db, six decayed by the emission of an alpha particle, and two underwent EC decay to ^{256}Rf, corresponding to a 78 ± 14% alpha decay branch and a 22 ± 14% EC branch. These values are in agreement with the observed 36 ± 12% EC branch found in the literature[15]. A weighted mean of the alpha branch from previous work and our current findings results in an alpha branch of 70 ± 11%. The six alpha decays correspond well to known alpha decay groups, with the alpha decays in chains 2, 3, 4, 6, and 8 in Table 1 belonging to the group at 9.01 MeV, and the alpha decay in chain 5 belonging to the group at 9.12 MeV. The half-life of these six ^{256}Db alpha events and the two EC events is $1.1^{+0.6}_{-0.3}$ s, consistent with the reported value of $1.6^{+0.5}_{-0.3}$ s[15].

The two spontaneous fissions observed in this experiment were the result of production of ^{256}Rf, the EC daughter of ^{256}Db. ^{256}Rf decays by SF with a branching of >98%[16, 17]. These SF decays were observed in decay chains 1 and 7 with measured energies of 148.0 and 174.3 MeV, respectively. No half-life was measured for ^{256}Rf because lifetimes were measured as the sum of the ^{256}Db and the ^{256}Rf lifetimes.

Six alpha decays of ^{252}Lr were observed as the granddaughter decay of ^{260}Bh. The half-life of these events is $0.27^{+0.18}_{-0.08}$ s. The ^{252}Lr alpha particles in chains 6 and 8 escaped the focal plane detector, registering 2.39 and 3.31 MeV, respectively. The 8.99 and 9.02 MeV decays in chains 3 and 4 fit well to the known alpha decay groups at 8.974 MeV and 9.018 MeV, respectively[15]. The remaining decays at 8.82 and 9.61 MeV have different energies than any group previously observed in the alpha decay of ^{252}Lr, and may represent new alpha lines. It is important to note that the highest energy decay, 9.61 MeV, is 0.5 MeV higher than that from the expected Q-value for this decay[11]. Careful examination of the data supports that it is a valid alpha decay of ^{252}Lr and a member of a ^{260}Bh decay chain, but at this time we are unable to explain this high energy further. No SF decays or alpha decays resembling ^{252}No were observed, supporting earlier claims [13] contending ^{252}Lr decays by alpha emission only.

^{248}Md, the alpha decay great-granddaughter of ^{260}Bh, was observed to decay both by electron capture to ^{248}Fm and through alpha emission to ^{244}Es in this experiment. Five of the six alpha decay chains passed through ^{248}Md. Three of these five events decayed by emission of alpha particles of 8.26, 8.46, and 8.13 MeV. The total half-life from the three alpha events is $12.5^{+14.8}_{-4.3}$ s, consistent with 7 ± 3 s [18]. This results in a 58 ± 20% alpha branch in contrast to the 20% branch reported in previous work, the apparent discrepancy may be due to the low counting statistics in our study. The ^{248}Md alpha energy of 8.46 MeV was observed to follow the 9.61 MeV decay of ^{252}Lr. These correlated high-energy transitions could be interpreted in terms of isomerism in ^{252}Lr and ^{248}Md, however, in the absence of data such as gamma spectra, we do not suggest any level schemes at this time. Two events correlating to the alpha decay of ^{248}Fm were observed in this work, following the EC decay of ^{248}Md. The two events registered alpha decay energies of 7.85 and 8.06 MeV. The half-life for this isotope cannot be determined directly because its lifetimes were measured as the sum of the ^{248}Md and ^{248}Fm lifetimes.

A random event correlation analysis was conducted for this work, analogous in method to the one described in the appendix of [28]. The maximum time of individual event consideration, Δt_{max}, was chosen as 35 seconds, a multiple of five times the longest literature value for the half-life of ^{248}Md. The focal plant event rates, R_α and N_{EVR} for the rate of alpha-like events and number of EVR-like events, respectively, were determined by integrating over their spectra. The total time of experimental data acquisition was 123,980 seconds. Alpha-like events were required to have energies between 7.5 - 11 MeV, and EVR-like events were required to have energies within the same energy gates as used in the online shutoff conditions. The number of randomly correlated decay chains expected was calculated by multiplying the Poisson probability of observing a certain number of alpha-like events per pixel within 35 seconds by the number of EVR-like events. The pixel size was defined as ±1.5 mm within a single strip. The number of random chains expected over the duration of the experiment from an EVR followed by two alpha-like events was $1.2 \cdot 10^{-3}$, and much lower for EVR-SF chains or EVRs followed by greater than two alpha-like events. Therefore, we conclude that the multiple sequential alpha decay chains observed in this work are true events and not random correlations.

We found the experimental magnetic rigidity of the EVRs to be 2.14 T·m and the corresponding charge state to be 7.8, very close to our predicted values. The total integrated beam dose was $1.7 \cdot 10^{17}$ ions. The measured cross section from these eight decay chains of ^{260}Bh is 59^{+29}_{-20} pb. This cross-section calculation includes a 97% efficiency for detection of a decay chain. We have defined these decay chains as an EVR correlated in time and position to a minimum of two full-energy alpha decays or an SF decay. This cross-section is nearly a factor of four greater than the theoretical prediction of 15 pb at an energy of approximately 202 MeV in the center-of-mass frame. W. J. Świątecki graciously provided us this prediction from the "Fusion By Diffusion" model[21, 22] after the experiment had been conducted. Because only one bombarding energy was run in this work, it is not known if 59^{+29}_{-20} pb is the peak of the ^{209}Bi(^{52}Cr,*n*) excitation function. Therefore, it would be instructive to continue this study by exploring the same reaction at additional energies to map the entire excitation function.

There is also evidence for the influence of the deformed N = 152 shell on the alpha decay energies in this region of the Bh isotopes. Among the N = 153-155 isotones, the N = 154 isotones possess the largest alpha decay energies as a result of decaying into the N = 152 shell. This value is approximately 150-340 keV greater than the N = 153 or N = 155 members' alpha decay energy. The N = 153-155 isotopes of Bh follow this trend as well, with the ^{260}Bh major alpha group decaying with an energy of 10.16 MeV,

^{261}Bh with 10.40 MeV, and the ground state isomer of ^{262}Bh with 10.06 MeV, respectively. In the absence of the N = 152 shell a smooth decrease in Q_α with increase in N is expected.

Investigating the decay properties of the next lightest Bh isotope, ^{259}Bh (N = 152), will provide more information about these decay energy trends. This isotope could be produced by using the ^{209}Bi(^{52}Cr,2*n*)^{259}Bh reaction. It is also possible that ^{259}Bh will exhibit unusual alpha stability, being at the N = 152 deformed shell and only one proton away from the Z = 108 shell. Studying ^{259}Bh as well as other even-N isotopes of Bh can also provide a better understanding of SF hindrance in this region.

In conclusion, we have observed eight decay chains and measured a cross section for the production of the new alpha-decaying transactinide isotope ^{260}Bh in the reaction ^{209}Bi(^{52}Cr,*n*). The decay properties of this isotope are similar to those calculated from systematics, with a half-life of 35^{+19}_{-9} ms and a dominant alpha group at 10.16 MeV. This result expands our knowledge of the decay behavior and the possible location of closed shells in this section of the chart of the nuclides.

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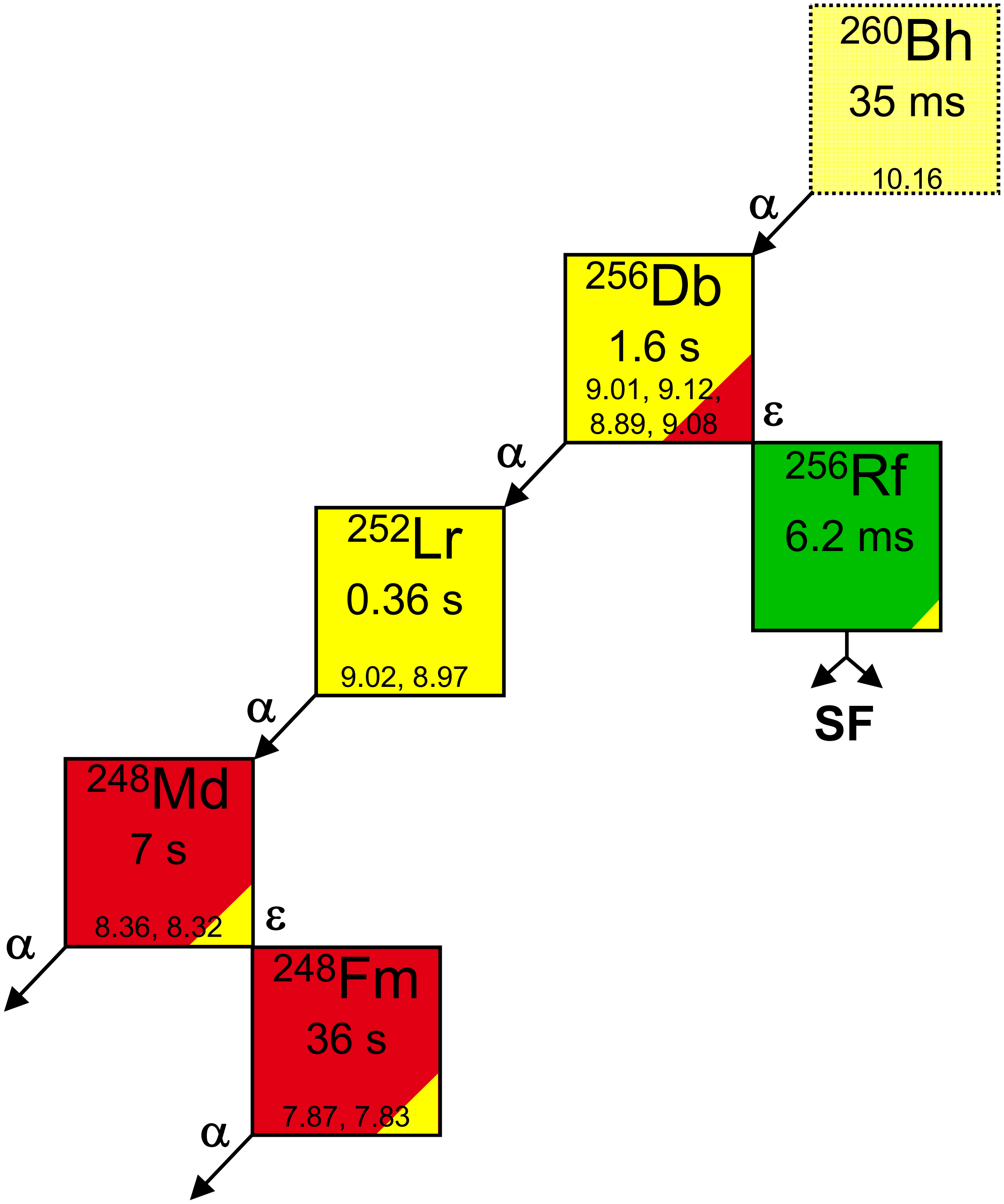

Figure 1: Decay properties of ^{260}Bh and its previously known daughter nuclides. [15-18]
Energies given in MeV. (color online)

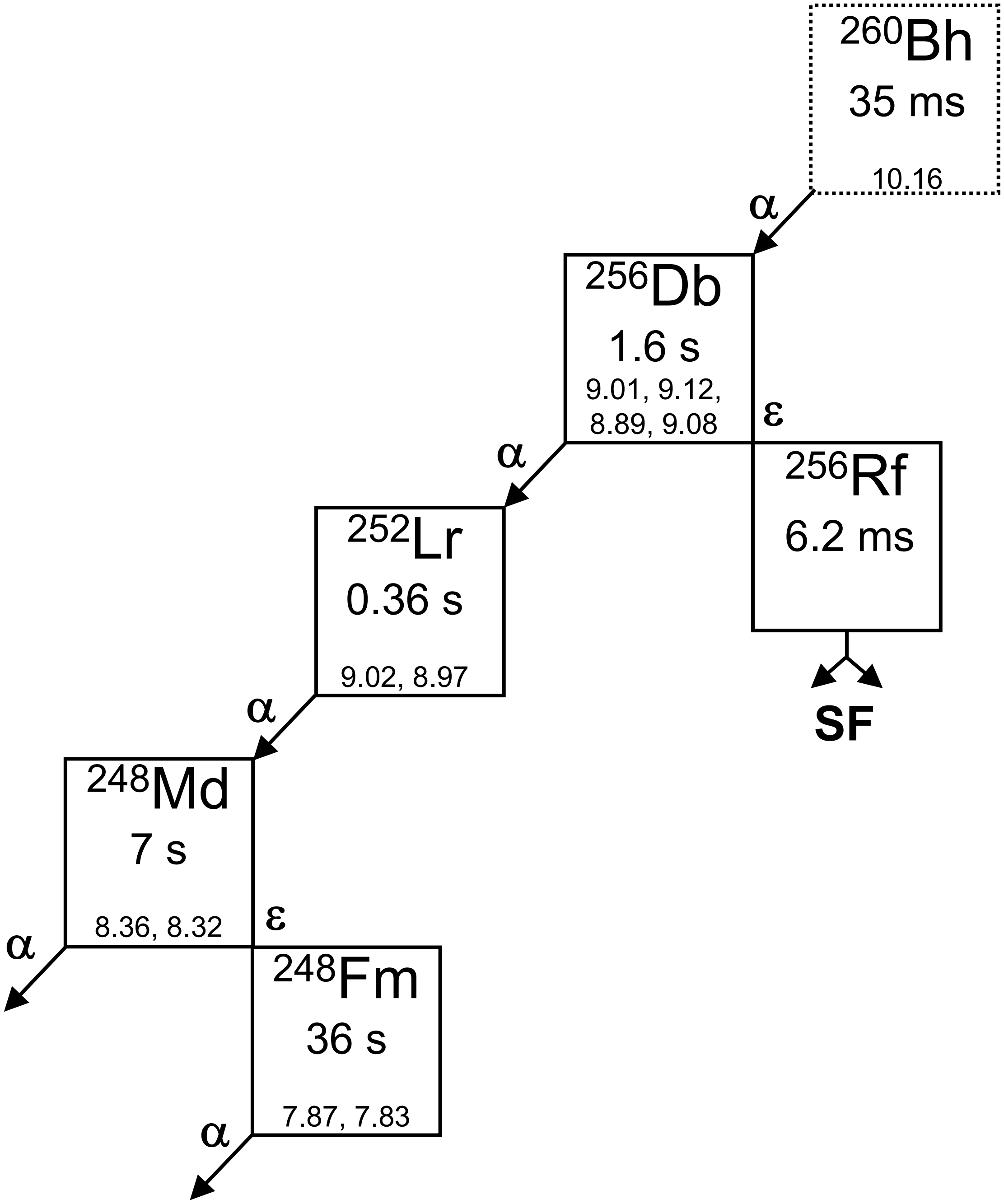

Figure 1: Decay properties of ^{260}Bh and its previously known daughter nuclides. [15-18]
Energies given in MeV. (black and white)

Event #	Strip #	E_{EVR} (MeV)	Position (mm)	Decay Energy (MeV)	Position (mm)	Lifetime	A,Z
1	24	18.0	-28.0	10.24	-28.8±0.3	0.044 s	260Bh
				EC			256Db
				148.0	-27.9±1.5	0.648 s	256Rf
2	38	14.9	9.0	10.17	8.6±0.3	0.003 s	260Bh
				9.03	8.2±0.3	0.380 s	256Db
				8.82 [0.36+(0.22)+8.24]	>0.0	0.043 s	252Lr
				8.26	8.9±0.3	36.460 s	248Md
3	32	15.2	9.1	10.17	8.6±0.3	0.135 s	260Bh
				9.06	8.9±0.3	0.316 s	256Db
				8.99	8.8±0.3	1.394 s	252Lr
				EC			248Md
				7.85	8.7±0.4	59.642 s	248Fm
4	21	16.5	28.4	escape, 0.77 [0.72+(0.05)]	28.4±3.9	0.045 s	260Bh
				9.02 [1.66+(0.11)+7.25]	>0.0	0.563 s	256Db
				9.02 [1.00+(0.06)+7.96]	>0.0	0.119 s	252Lr
5	20	16.5	7.0	10.08	7.4±0.3	0.050 s	260Bh
				9.19	7.1±0.3	0.761 s	256Db
				9.61 [1.20+8.41]	8.1±2.3	0.195 s	252Lr
				8.46	7.2±0.3	1.672 s	248Md
6	34	17.9	20.1	10.19	19.7±0.3	0.015 s	260Bh
				9.04	19.4±0.3	4.600 s	256Db
				escape, 2.38	15.5±1.2	0.540 s	252Lr
				8.13 [0.80+(0.20)+7.13]	>0.0	15.783 s	248Md
7	36	18.1	3.6	10.13 [1.16+8.97]	1.2±2.4	0.080 s	260Bh
				EC			256Db
				174.3	4.0±1.5	1.120 s	256Rf
8	14	16.3	7.5	10.03	7.4±0.3	0.037 s	260Bh
				9.03	8.0±0.3	4.323 s	256Db
				escape, 3.31	7.6±0.9	0.065 s	252Lr
				EC			248Md
				8.06 [1.33+6.73]	7.9±2.2	25.171 s	248Fm

Table 1: Observed ^{260}Bh decay chains. Reconstructed energies are listed in square brackets with the focal plane energy listed first, followed by the calculated energy from a missing signal from either the top or bottom of the strip in parentheses, ending with the energy deposited in the upstream detector (see text). Positions listed as greater or less than 0.0 mm indicate an event missing an energy signal. Boldface type indicates decay was observed during a beam-off interval. Lifetimes of decays following EC are the sum of the two lifetimes.