

Supplementary Information for:
A pathway towards decentralized studies of
radioactive post-lead elements and their
applications in beyond standard model physics

Moritz Pascal Reiter^{1*}, Kriti Mahajan^{2,3}, Meetika Narang^{4,5},
Carsten Zülch⁶, Timo Dickel^{2,4}, Daler Amanbayev^{2,4},
Robert Berger⁶, Julian Bergmann², Agnieszka Bukowicka¹,
Mariam Fadel⁶, Tayemar Fowler-Davies^{1,4}, Zhuang Ge⁴,
Simeon Gloeckner⁴, Gabriella Kripkó-Koncz^{1,2},
Nasser Kalantar-Nayestanaki⁵, Cameron Merron¹,
David J. Morrissey^{4,7}, Wolfgang R. Plaß^{2,4},
Christoph Scheidenberger^{2,3,4}, Makar Simonov^{2,3},
Nazarena Tortorelli^{4,8}, Jiajun Yu⁴, Alexandra Zadvornaya^{1,2},
Jianwei Zhao⁴

¹School of Physics and Astronomy, University of Edinburgh, Edinburgh,
EH9 3FD, United Kingdom.

²II. Physikalisches Institut, Justus-Liebig Universität Giessen, Giessen,
35392, Germany.

³Helmholtz Research Academy Hesse for FAIR (HFHF), GSI Helmholtz
Center for Heavy Ion Research, Campus Gießen, Giessen, 35392,
Germany.

⁴GSI Helmholtz Centre for Heavy Ion Research GmbH, Darmstadt,
64278, Germany.

⁵Nuclear Energy Group ESRIG, University of Groningen, Groningen,
9747 AA, The Netherlands.

⁶Department of Chemistry, Philipps-Universität Marburg, Marburg,
35032, Germany.

⁷Department of Chemistry, Michigan State University, East Lansing, MI
48824, United States.

⁸Faculty of Physics, Ludwig-Maximilians-Universität München,
Garching, 85748, Germany.

*Corresponding author(s). E-mail(s): mreiter@ed.ac.uk;

Supplementary Note 1: Validation of quantum-chemical calculations

Supplementary Table 1 and Supplementary Table 2, benchmark the electronic structure approach used in this work (RECP-ROHF-UCCSD(T)) against available literature data for ionization energies, electron affinities and selected dissociation energies of Pb-, Po- and Ra-containing species. For Ra and RaF, the computed ionization energies agree closely with experiment, supporting the accuracy of the method for the key charge states involved in the fluorination reactions. For Pb and Po, larger deviations are observed for some ionization energies, consistent with the more pronounced multi-reference character of their low-lying electronic states and remaining spin-orbit effects not explicitly included in the present treatment. For Po, the electron affinity is reproduced within the range of reported theoretical values. Overall, these benchmarks support the use of the calculations to interpret the experimentally observed reaction channels and trends, while absolute energies for Pb/Po should be viewed with correspondingly larger uncertainty.

Supplementary Table 1 Electronic ionization energies (IE), electron affinities (EA) and dissociation energies (DE) of the various F, OH and SF_{5,6} species computed adiabatically on the level of ROHF-UCCSD(T) with and without frozen orbitals are given with literature values if present.

	Lit.		ROHF-UCCSD(T)-frozen-core	ROHF-UCCSD(T)
IE(F)	17.4	[1]	17.4	17.4
EA(F)	3.40	[2]	3.38	3.39
IE(OH)	13.0	[3]	13.0	13.0
EA(OH)	1.83	[4]	1.79	1.80
DE(H ₂ O → H + OH)	5.10	[5]	5.42	5.44
IE(SF ₆)	15.1	[6]	16.0	16.0
IE(SF ₅)	9.60	[7]	9.84	9.78
DE(SF ₆ → SF ₅ ⁺ + F ⁻)			11.1	11.2
DE(SF ₆ → SF ₅ + F)			4.63	4.82

Supplementary Table 2 Electronic ionization energies (IE), electron affinities (EA) and dissociation energies (DE) of relevant systems including Pb, Po or Ra computed adiabatically on the level of ROHF-UCCSD(T) are given with selected experimental (Lit. Exp.) and theoretical (Lit. Theo.) data from literature.

	Lit. Exp.		Lit. Theo.	ROHF-UCCSD(T)
IE(Pb)	7.42 [8]		7.04 [9]	6.87
IE(Pb ⁺)	15.0 [10]		14.3 [9]	13.4
IE(PbF)	7.55 [11]		7.44 [12]	6.72
DE(PbF ⁺ → Pb + F ⁺)			15.1 [13]	13.0
DE(PbF ⁺ → Pb ⁺ + F)			4.72 [13]	2.54
DE(PbOH ⁺ → Pb + OH ⁺)				8.10
DE(PbOH ⁺ → Pb ⁺ + OH)				1.98
IE(Po)	8.42 [14]		8.43 [15]	8.08
IE(Po [4z])	8.42 [14]		8.43 [15]	8.18
IE(Po [5z])	8.42 [14]		8.43 [15]	8.22
IE(Po ⁺)			19.4 [16]	17.8
IE(Po ⁺ [4z])			19.4 [16]	17.9
IE(Po ⁺ [5z])			19.4 [16]	17.9
EA(Po)		1.469-1.9 [15, 17] ^a		1.79
EA(Po [4z])		1.469-1.9 [15, 17] ^a		1.88
EA(Po [5z])		1.469-1.9 [15, 17] ^a		1.91
IE(PoF)				8.50
IE(PoF ⁺)				17.3
IE(PoF ⁺ [4z])				17.3
DE(PoF ⁺ → Po + F ⁺)				12.0
DE(PoF ⁺ → Po ⁺ + F)				2.77
DE(PoOH ⁺ → Po + OH ⁺)				6.66
DE(PoOH ⁺ → Po ⁺ + OH)				1.76
IE(Ra)	5.28 [18]		5.29 [19]	5.23
IE(Ra ⁺)	10.1 [20]		10.1 [21]	10.1
IE(RaF)	4.97 [22]		4.97 [22]	4.95
DE(RaF ⁺ → Ra + F ⁺)				17.9
DE(RaF ⁺ → Ra ⁺ + F)			5.59 [23]	5.74
DE(RaOH ⁺ → Ra + OH ⁺)				12.4
DE(RaOH ⁺ → Ra ⁺ + OH)				4.59

^aUsed ECP was assessed against EA(Po)=1.90 eV [17]. A more recent computation on the level of 4c+CCSD(T)+Breit+QED predicts 1.47 eV [15].

Supplementary References

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