

**Supplementary Information: Effect of *f*-element Complexation on the Radiolysis of 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester (HEH[EHP])**

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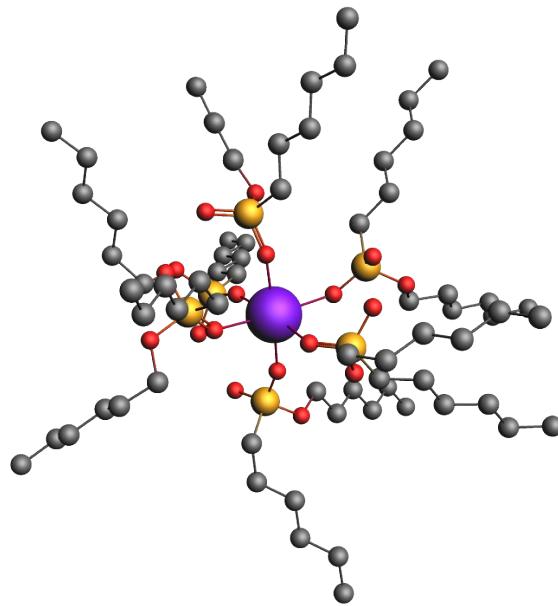
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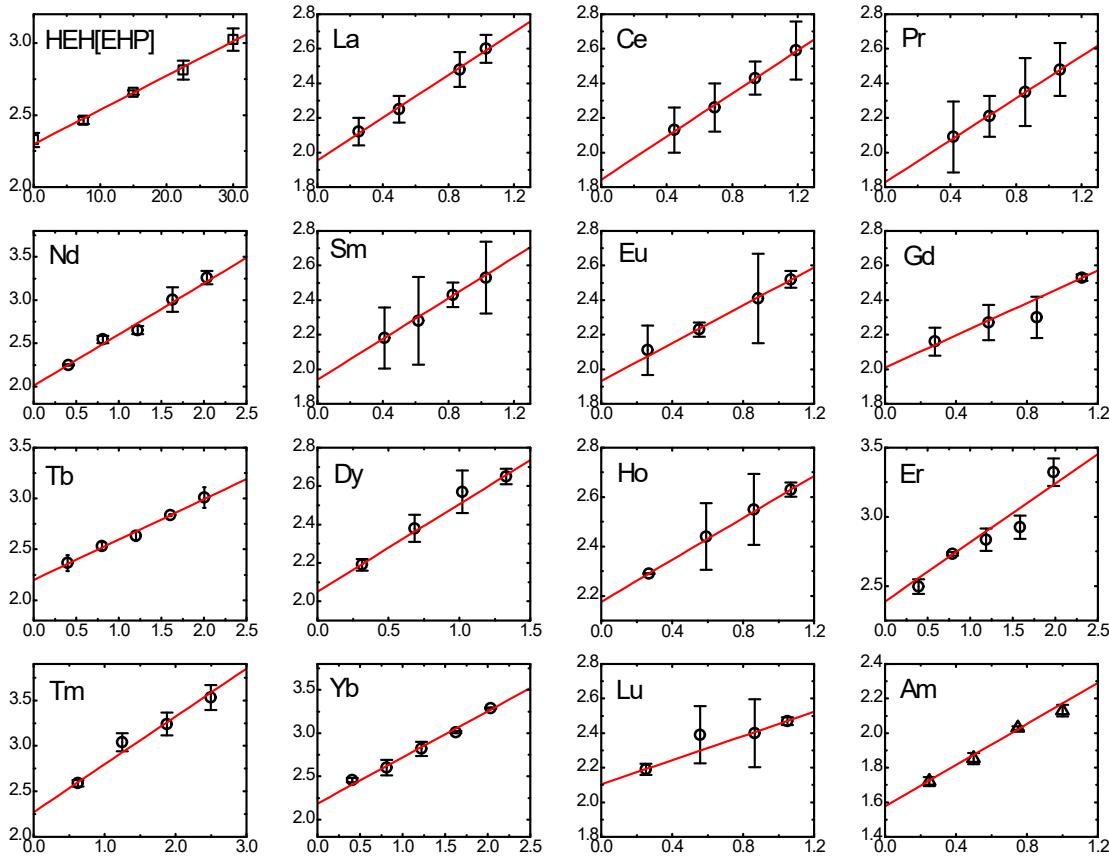
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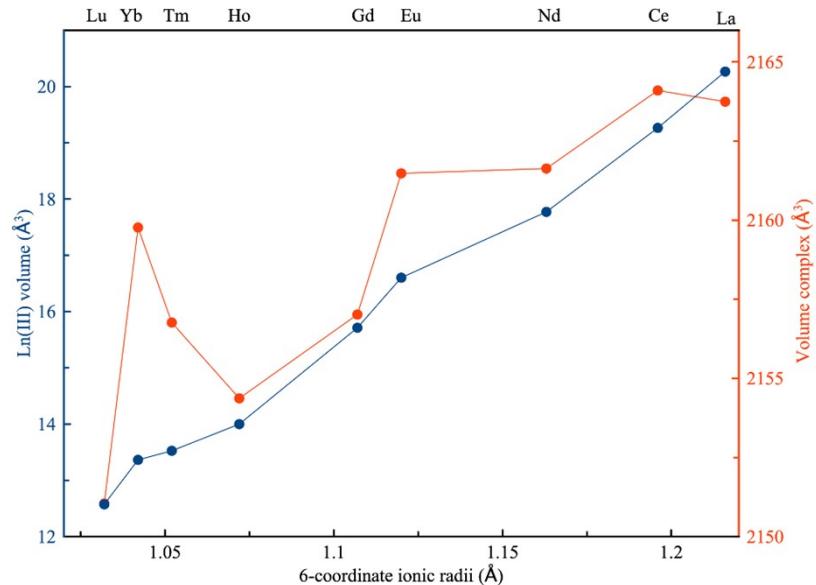
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**Fig. S1.** Computational model structure used to evaluate the bonding properties of the *f*-element coordinated HEH[EHP] complexes.



**Fig. S2.** Second-order rate coefficient determinations for non-complexed HEH[EHP], Ln-complexed HEH[EHP] (La–Lu), and americium complexed HEH[EHP]. For all plots: y-axes ( $10^{-8} \text{ s}^{-1}$ ), x-axes [Species] (mM). Slope values correspond to second-order rate coefficients as detailed in main text, **Table 1**.



**Fig. S3.** Correlation between 6-coordinate ionic radii and calculated molecular (red) and Ln(III) (blue) volumes from the topological analysis.