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Nitrogen-rich complexes of p-block elements: Highly endothermic polytetrazolates and polyazides

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Our aim is to stabilise and control the reactivity of N-rich coordination compounds. Upon decomposition, N-rich compounds generate mainly N_2 - ideal for smokeless, CO_2 -free, “green” energetic materials as replacement for conventional propellants, explosives & pyrotechnics.¹ Low barriers toward decomposition pose major challenges in their preparation and characterisation. Here we discuss $E(Y)_n$ complexes with E = group 13, 14 or 15 coordination centre in low or high oxidation state, Y = N-rich ligand, n = 2-6. We apply synthetic methods novel to energetic chemistry, which involves a combination of hypercoordination, bulky counterions, and ligand exchanges, to achieve the synthesis and isolation of new classes of complexes as candidates for efficient & controllable energy storage.⁴ These include Lewis base adducts,^{3,6} homoleptic azido complexes,^{2,5} covalent, binary azides $E(N_3)_n$,³ the first homoleptic p-block element tetrazolates $E(T)_6^{2-}$ and $E(T)_3^-$ (E = Si - Sn, T = N_4CH , Fig. 1). These complexes have a unique chemistry – reactions with nitriles and phosphines afford unusual poly(tetrazolato) and poly(phosphiniminato) complexes. Syntheses, reactivity, structures, and thermal and spectroscopic properties of the new species, including the application of fast time-resolved IR spectroscopy to study photoreactivity, will be described.

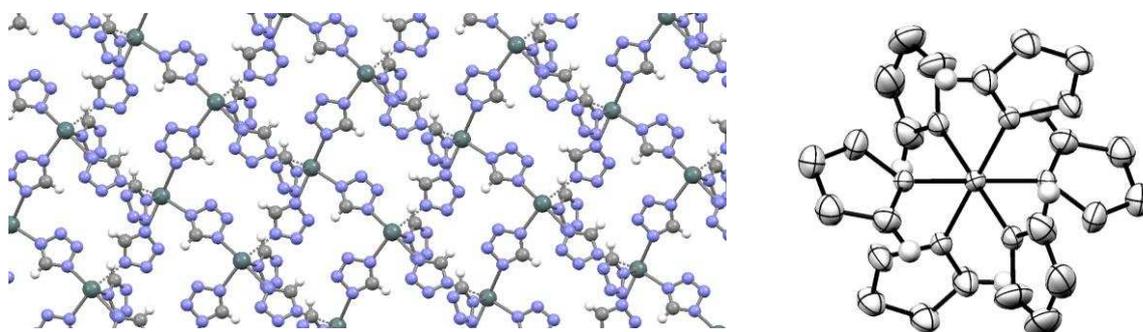


Figure 1 Tetrazole-based N-rich coordination networks and complexes: left $\{Sn(CHN_4)_3\}^-_\infty$ (blue N, turquoise Sn, white H), right $[Si(\kappa-N(1)-N_4CH)_6]^{2-}$ (thermal ellipsoid plot, 50%).

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