Supporting Information

Unveiling the potential of redox chemistry to form size tunable, high index silicon particles

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Figure S1. Molecular structure of bis(N,N'-diisopropylbutylamidinate) dichlorosilane. Referred to as $SiCl_2[BuC(N^iPr)_2]_2$ throughout this publication.



Figure S2. Particles produced when excess sodium silicide is reacted with $SiCl_2[BuC(N^iPr)_2]_2$ in toluene (1.5 : 1 molar ratio). Scale bar represents 1 μ m.



Figure S3. TEM image of particles formed when 5 molar eq. $SiCl_2[BuC(N^iPr)_2]_2$ is reacted with 1 molar eq. Na_4Si_4 in toluene for 16 hours. Scale bar represents 1 μ m.



Figure S4. Powder X-ray diffractograms of particles produced when sodium silicide is reacted with $SiCl_2[BuC(NPr)_2]_2$ in toluene for 16 hours with a (blue) 1:1:5 molar ratio, (purple) 1:2 ratio, (pink) 1:3 ratio, and (red) 1:4 ratio.



Figure S5. Additional experiments varying the quantity of Si (IV) species and ligand separately to a fixed quantity of Na_4Si_4 in toluene and reacted for 16 hours. An overall reaction concentration of 20 mmol was maintained in all cases.



Figure S6. Growth experiments stopped early, (a) after 1 hour and (b) after 4 hours using 2 eq $SiCl_2[BuC(N^iPr)_2]_2$ with 1 eq. Na_4Si_4 in toluene. Scale bars represent 200 nm.



Figure S7. Optical image (a) and ToF-SIMS images of particles formed using $n=2 \text{ eq. SiCl}_2[BuC(N^iPr)_2]_2$ corresponding to (b) m/z peak at 255, (c) m/z peak at 283, (d) m/z peak for Au⁻.



Figure S8. ATR-FTIR spectroscopy of particles produced when sodium silicide is reacted with $SiCl_2[BuC(N^iPr)_2]_2$ in toluene for 16 hours with a (black) 1:1, (blue) 1:1:5, (purple) 1:2, (pink) 1:3, and (red) 1:4 molar ratio.

Molar Ratio	mg SiCl ₂ [BuC(N ⁱ Pr) ₂] ₂	mg Na₄Si₄
1:1	37.4	16.5
1:1.5	44.3	13.2
1:2	49.9	11.0
1:3	54.5	9.0
1:4	59.9	6.6
1:5	62.4	5.5

Table S1. Actual precursor quantities used to maintain 20 mmol concentration in 8 mL toluene.



Figure S9. The distribution of measured indices computed from 170 interference pattern images, captured by a single particle formed using n=4 eq. SiCl₂[BuC(NⁱPr)₂]₂.