Tinning the Carbon: Hydrostannanes Strikes Back

Pavel Rublev,[a,‡] Nikolay V. Tkachenko,[a,‡] Anton S. Pozdeev,[a] and Alexander I. Boldyrev*[a]

a. Department of Chemistry and Biochemistry, Utah State University, Logan, Utah 84322, United States. E-mail: a.i.boldyrev@usu.edu;
‡ Those authors contributed equally

Supporting Information

Contents

S1. Population size in Coalescence Kick simulations

Table S1. *Population size in Coalescence Kick simulations*

Stoichiometry	Population Size	Stoichiometry	Population Size
Sn_2H	750	Sn_3H_2	1000
Sn_2H_2	1000	Sn_3H_3	1000
Sn_2H_3	1000	Sn_3H_4	2000
Sn_2H_4	1000	Sn_3H_5	3000
Sn_2H_5	2000	Sn_3H_6	5000
Sn_2H_6	3000	Sn_3H_7	7000
Sn_3H	1000	Sn_3H_8	10000

S2. Example of SA-MD Global Optimization execution script

```
%md

Cell Cube 10 Spring 20
# Cell cube creates cubic wall around center of mass
# Spring is an elastic constant (kJ/mol) within quadratic potential
Minimize LBFGS Steps 5 Noise 0.45
# L-BFGS minimization is only needed for generating adequate initial
# structure. Noise generates random atomic displacements

# The next block is an MD Block
Initvel 1500_K
# Velocities generation for concrete temperature
Thermostat NHC Timecon 1_fs Massive Chain 5 MTS 5 # Thermostat settings
Timestep 0.5_fs # Integrand step length
Run 500 # How much steps in MD trajectory

# The next block is an annealing block

Minimize Anneal Steps 3000 TempConv 1250_K Noise 0.1
# TempConv is a convergence temperature
# Noise is a random atomic displacement
Dump Position Format XYZ Stride 0 Filename "Geometry_1500.xyz"
# Put the final geometry in the new file

Initvel 1250_K # New random velocities for the next temperature step
Thermostat NHC 1250 Timecon 1_fs Massive Chain 4 MTS 5
Run 400
Timestep 0.5_fs
Minimize Anneal Steps 3000 TempConv 1000_K Noise 0.1
Dump Position Format XYZ Stride 0 Filename "Geometry_1250.xyz"
```

```
Initvel 1000_K
Thermostat NHC 1000 Timecon 2_fs Massive Chain 4 MTS 5
Run 400
Timestep 0.5_fs
Minimize Anneal Steps 3000 TempConv 800_K Noise 0.1
Dump Position Format XYZ Stride 0 Filename "Geometry_1000.xyz"

Initvel 800_K
Thermostat NHC 800 Timecon 2_fs Massive Chain 3 MTS 5
Timestep 0.5_fs
Run 300
Minimize Anneal Steps 3000 TempConv 600_K Noise 0.1
Dump Position Format XYZ Stride 0 Filename "Geometry_800.xyz"

Initvel 600_K
Thermostat NHC 600 Timecon 2_fs Massive Chain 3 MTS 5
Timestep 0.5_fs
Run 300 CenterCOM
Minimize Anneal Steps 3000 TempConv 300_K Noise 0.1
Dump Position Format XYZ Stride 0 Filename "Geometry_600.xyz"

Initvel 300_K
Thermostat NHC 300 Timecon 5_fs
Timestep 0.5_fs
Run 200
Minimize Anneal Steps 3000 TempConv 100_K Noise 0.05
Dump Position Format XYZ Stride 0 Filename "Geometry_300.xyz"

Initvel 100_K
Thermostat NHC 100 Timecon 5_fs
Timestep 0.5_fs
Run 100
Minimize Anneal Steps 3000 TempConv 50_K Noise 0.05
Dump Position Format XYZ Stride 0 Filename "Geometry_100.xyz"

Initvel 50_K
Thermostat NHC 50 Timecon 5_fs
Timestep 0.5_fs
Run 100
Minimize Anneal Steps 3000 TempConv 5_K Noise 0.025
Dump Position Format XYZ Stride 0 Filename "Geometry_50.xyz"

Initvel 5_K
Thermostat NHC 5 Timecon 10_fs
Timestep 0.5_fs
Run 100
Minimize Anneal Steps 3000 TempConv 1_K Noise 0.01
Dump Position Format XYZ Stride 0 Filename "Geometry_5.xyz"

# Final L-BFGS geometry optimization
Minimize LBFGS Steps 3000 StepLimit 0.1
Dump Position Format XYZ Stride 0 Filename "Geometry_Final.xyz"


End
```

S3. Examples of SA-MD Global Optimization trajectory

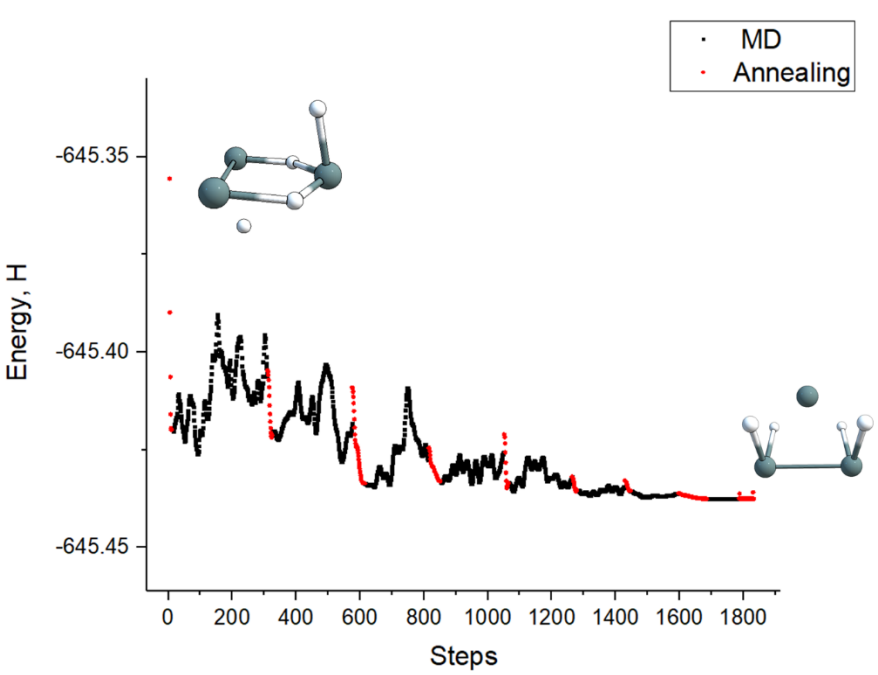


Figure S3. *Global Optimization trajectory of $Sn_3H_4^S$: red dots correspond to simulated annealing blocks, black dots correspond to MD blocks*

S4. Execution performance of global optimization techniques

In this section we compared the execution performance of used global optimization techniques toward optimization of Sn_3H_2, Sn_3H_4, and Sn_3H_6 stoichiometries. It is worth to mention, this performance overview carries only an approximate character. Due to completely different program codes, linked MPI packages, OpenMP implementation, and architecture of computational clusters, timings and performance (in sense of core-hours) have a low quantitative meaning. For example, the absence of AVX-512 instructions steadily reduces performance of the Gaussian16 suite.

Table S4. *An overview of execution performance. All calculations were performed using one computational node and different number of cores: 8 cores for CK calculations, 32 cores for SA-MC, and 32 cores for SA-MD calculations.*

Stoichiometries	CK, core-hours	SA-MC, core-hours	SA-MD, core-hours
Sn_3H_2	111.84	90.61	122.88
Sn_3H_4	452.08	149.51	153.6
Sn_3H_6	1540.88	244.03	225.28

S5. Spin-state energy difference

Table S5. *Spin-state energy difference. Energy differences were calculated on U-RSX-PBE-QIDH / [SARC/def2]-ZORA-TZVPP level of theory. Global minimum geometries were obtained through SA-MD minimization.*

Stoichiometries	$E^T - E^S$, kcal/mol	Stoichiometries	$E^D - E^Q$, kcal/mol
Sn_2H_2	24.74	Sn_2H	18.22
Sn_2H_4	18.38	Sn_2H_3	21.53
Sn_2H_6	92.20	Sn_2H_5	79.69
Sn_3H_2	24.31	Sn_3H	26.50
Sn_3H_4	35.29	Sn_3H_3	17.51
Sn_3H_6	45.13	Sn_3H_5	63.31
Sn_3H_8	38.27	Sn_3H_7	78.18

S6. Sn_2H_x and Sn_3H_y global minimum geometries (XYZ coordinates)

Table S6. *Sn_2H_x and Sn_3H_y global minimum geometries*

Stoichiometry^{Multiplicity}	XYZ coordinates			
Sn_2H^D	Sn	0.455809605	1.057965691	0.602170708
	Sn	-0.722583957	-0.694937180	-1.211623768
	H	0.644274352	0.755871488	-1.364646940
Sn_2H^Q	Sn	-0.855855565	1.083036929	-2.116552219
	Sn	0.068411394	-1.379685046	-3.283676308
	H	0.229621849	2.249602849	-2.939796990
$Sn_2H_2^S$	Sn	-1.168891835	0.539021926	0.876332227
	Sn	-0.641640985	-1.125730054	-1.293270526
	H	0.460578984	0.051558786	-0.141380734
	H	-1.135246164	-1.424150659	0.603020033
$Sn_2H_2^T$	Sn	-1.383802194	-0.288377937	-0.992194993
	Sn	-0.092174588	-0.019724261	-3.623212588
	H	1.489893209	0.547694292	-2.997176584
	H	-0.100016587	-1.316809165	-2.124449663
$Sn_2H_3^D$	Sn	0.728392883	-0.036499175	1.224101472
	Sn	-1.151746714	-1.117196581	3.523830706
	H	-1.001515628	0.248097254	2.095312046
	H	0.519606960	-1.484688730	2.525223968
	H	1.457662499	0.930487232	2.544532808
$Sn_2H_3^Q$	Sn	-1.180009548	1.194370850	-1.937950877
	Sn	0.897023206	-0.619859515	-2.573954891
	H	0.273488407	-2.150436022	-3.130868960
	H	1.937913482	0.005136936	-3.826139109
	H	-2.359361077	1.705374011	-3.142099579
$Sn_2H_4^S$	Sn	-1.136889863	0.737481406	2.668745389
	Sn	1.993939611	0.824792450	3.070611411
	H	0.559451265	1.167654893	1.766359678
	H	1.698843532	2.504786224	3.624959880
	H	-0.841946562	-0.942527657	2.114360621
	H	0.297603017	0.394512684	3.972963021

$Sn_2H_4^T$	Sn	0.127966896	0.657789334	-1.247555190
	Sn	-0.805856238	-0.287436401	-3.763785876
	H	-0.800836128	-2.029903416	-3.853919560
	H	-2.461394049	0.194743798	-3.993469026
	H	-1.099835032	0.561821053	-0.011842701
	H	1.439375417	-0.341698801	-0.693518192
$Sn_2H_5^D$	Sn	2.108958485	-1.014456080	-1.267037167
	Sn	-0.229803067	-0.594680124	0.258604416
	H	0.043847149	0.570262847	1.519175959
	H	1.822269561	-2.224955236	-2.453422332
	H	-0.704566315	-2.051903717	1.078808485
	H	3.445774704	-1.514575112	-0.300436881
	H	2.546319482	0.437707423	-2.076492481
$Sn_2H_5^Q$ (Dissociated)	Sn	0.774645435	-1.544662833	-2.444312283
	Sn	-1.467118107	1.389764887	-2.708439838
	H	-0.198792543	2.006288709	-1.705242984
	H	-0.765551939	0.237589885	-3.829864688
	H	2.317545508	-0.860851791	-2.866044948
	H	0.811915247	-2.881030083	-1.314928960
	H	-2.149636600	2.680153225	-3.657526299
$Sn_2H_6^S$	Sn	-1.728288171	2.399635986	-1.461194687
	Sn	-1.667016244	-0.290618532	-2.230362443
	H	-1.877115181	-1.321613474	-0.870779733
	H	-1.515405161	3.431417204	-2.819740921
	H	-0.477773966	2.745348689	-0.333290506
	H	-0.165803436	-0.685573201	-2.969086987
	H	-3.230785334	2.794044358	-0.724772458
	H	-2.919312508	-0.636741030	-3.356170266
$Sn_2H_6^T$ (Dissociated)	Sn	-0.779116539	0.688312879	-0.774565165
	Sn	0.355316694	-1.036894448	-5.550831415
	H	0.644980905	-1.868181247	-4.059873504
	H	-0.789551875	0.234594764	-5.288938857
	H	0.450175257	1.071092038	0.380994843
	H	-2.221560436	1.547762315	-0.357507963
	H	1.842826236	-0.351852463	-6.107688662
	H	-0.254997842	1.203998778	-2.342510081
Sn_3H^D	Sn	-0.260576098	1.528499565	-0.072908291
	Sn	-1.835210267	0.420616079	2.049172118
	Sn	0.292316021	-1.176097144	1.011428514
	H	0.972170344	0.047782499	-0.345492340
Sn_3H^Q	Sn	-0.878904482	-1.911970184	1.678454202
	Sn	-0.445674209	0.413480343	3.297715793
	Sn	0.562222516	0.435443194	0.465397541
	H	2.180251242	-0.181949658	0.941862921
$Sn_3H_2^S$	Sn	0.727902074	0.878723511	-0.293740091
	Sn	-2.185837247	1.386181862	-0.703742448
	Sn	-1.374888301	-1.155232380	0.291785180
	H	0.466278130	-1.095818737	0.403470065
	H	-0.633954657	2.351845744	-0.955673706

$Sn_3H_2^T$	Sn	1.819015600	-0.495908174	0.339710660
	Sn	-0.496589471	-0.098972894	2.437990706
	Sn	-1.106874866	-1.148212326	-0.184995000
	H	1.451281803	0.090712244	2.188339902
	H	1.497419838	1.203845124	-0.148345150
$Sn_3H_3^D$	Sn	1.270036351	-2.117935150	-0.084955123
	Sn	2.122325053	0.285600977	1.365933310
	Sn	-0.784830911	0.169562463	0.292319488
	H	-1.129624630	-0.724233903	1.819455116
	H	2.016996117	-1.664211748	1.695523326
	H	2.656598020	-0.694380639	-0.284775118
$Sn_3H_3^Q$	Sn	0.035456475	-1.176831615	-1.600753075
	Sn	-2.130027714	-1.997140502	0.416598938
	Sn	0.590293838	-1.180064496	1.597191150
	H	1.391981093	-1.005300651	-0.200049811
	H	1.460122339	-2.737062241	1.688362147
	H	0.003607367	0.608344259	-1.631361242
$Sn_3H_4^S$	Sn	1.270036351	-2.117935150	-0.084955123
	Sn	2.122325053	0.285600977	1.365933310
	Sn	-0.784830911	0.169562463	0.292319488
	H	-1.129624630	-0.724233903	1.819455116
	H	2.016996117	-1.664211748	1.695523326
	H	2.656598020	-0.694380639	-0.284775118
$Sn_3H_4^T$	Sn	1.108813886	1.446263376	-0.329519839
	Sn	-0.865860449	-0.760226034	0.227071069
	Sn	1.103240437	-1.473085089	-2.047657010
	H	-0.011258120	2.296339706	-1.432799959
	H	-0.158512626	-1.718899669	1.544412204
	H	1.630671826	0.404869612	-1.932680241
	H	2.473710033	-1.787198681	-0.943286883
$Sn_3H_5^D$	Sn	-0.103492749	2.509491813	-0.686055649
	Sn	1.650306233	0.143518363	-0.855524732
	Sn	-0.492812347	-0.407667952	1.096992949
	H	3.259921620	0.083209855	-0.226019295
	H	0.610464498	0.388460135	2.263915851
	H	-1.046873538	1.412445316	0.631283286
	H	1.748606235	-0.576593089	-2.425105841
	H	-1.105090839	1.650890808	-1.898576560
$Sn_3H_5^Q$ (Dissociated)	Sn	1.362448749	1.032395524	-1.640388479
	Sn	-0.898968687	-0.220771586	-0.363625744
	Sn	1.261027381	0.198235221	2.841808268
	H	2.353836133	0.656509565	1.596930720
	H	1.446891161	-1.453556453	3.250318839
	H	-0.337858978	0.476167676	2.227550498
	H	-0.026901746	-1.617035088	0.367292834
	H	1.484759519	1.180068172	4.234454243

$Sn_3H_6^S$	Sn	-0.323580236	-0.111673417	-2.626071896
	Sn	-0.652287545	2.082178206	-0.359965320
	Sn	0.669314319	0.156202929	1.395368646
	H	2.160909479	0.725530774	2.061677337
	H	-1.489182075	0.479880694	-1.145407278
	H	-1.139882918	1.147531107	-3.611278590
	H	1.061182909	-1.316509864	0.566479028
	H	-0.286243290	-0.318564374	2.757372070
	H	0.594368358	1.382623944	-1.717372998
$Sn_3H_6^T$ (Dissociated)	Sn	-0.715472411	-1.943101506	0.840735746
	Sn	-1.131902118	1.927526844	1.445634563
	Sn	-2.013906575	-0.819382457	-1.373666977
	H	-1.402026774	-2.676676814	2.265020430
	H	-1.122494529	0.608767023	2.555667038
	H	-2.461102031	1.688007011	0.373064190
	H	-1.300727586	3.417819859	2.286620150
	H	0.942270444	-1.700751221	1.327553225
	H	0.327397818	1.906303351	0.546405865
$Sn_3H_7^D$	Sn	0.661768027	4.074161993	-0.510711951
	Sn	-0.593782772	-0.435131990	-0.382228647
	Sn	1.529594229	1.392030302	-0.788310329
	H	-0.114643601	-2.046025853	-0.747962067
	H	-1.170184845	-0.407504601	1.242382033
	H	-0.853815257	4.243992686	-1.309049970
	H	-1.903238088	-0.027864298	-1.421626047
	H	0.464327364	4.540900523	1.136248753
	H	2.735366692	1.077816683	0.432035750
	H	1.774208250	5.166424554	-1.236778526
$Sn_3H_7^Q$ (Dissociated)	Sn	-2.432827335	0.120083583	-2.377562502
	Sn	-0.448026756	2.126667603	0.914190027
	Sn	0.676439113	-0.421510797	0.300996349
	H	0.102420465	2.636225571	2.489587556
	H	-0.370287157	-1.198846089	-0.875022261
	H	-3.578664106	0.716442319	-3.539917304
	H	-3.271406296	-0.306746048	-0.923001679
	H	0.728280821	-1.521690686	1.658131219
	H	-2.189170211	2.047320863	1.004251772
	H	-1.336298879	1.414477825	-1.977454591
$Sn_3H_8^S$	Sn	-2.687690496	-2.412720421	2.718530313
	Sn	0.955669900	0.098283015	1.210194954
	Sn	-0.502040706	-2.286437453	0.963697809
	H	-0.044798586	1.454579541	0.868064484
	H	0.542537991	-3.626120429	1.257713639
	H	1.552775058	0.275470758	2.813238049
	H	2.295768526	0.136872123	0.133429393
	H	-2.158175246	-2.305613076	4.351112096
	H	-3.561441767	-3.882577720	2.536077729
	H	-1.060894032	-2.449363123	-0.658991142
	H	-3.769911642	-1.108372216	2.427231678

$Sn_3H_8^T$ (Dissociated)	Sn	-2.444523113	0.404854341	-3.215726260
	Sn	1.583919828	-1.145546368	-3.434174691
	Sn	-0.514459609	2.485044093	-3.011197700
	H	0.193223153	-1.155257555	-4.446044087
	H	1.994056262	-2.751107061	-2.994194202
	H	2.894042954	-0.408906845	-4.259762082
	H	1.215773522	-0.239452373	-2.021749508
	H	-4.010806247	0.623653704	-2.452054890
	H	-1.175558353	3.995052107	-3.512309903
	H	0.095783869	2.625154517	-1.406115160
	H	0.770130522	2.034099282	-4.064713158

S7. AdNDP analysis of Sn_2H_x and Sn_3H_y global minimum structures

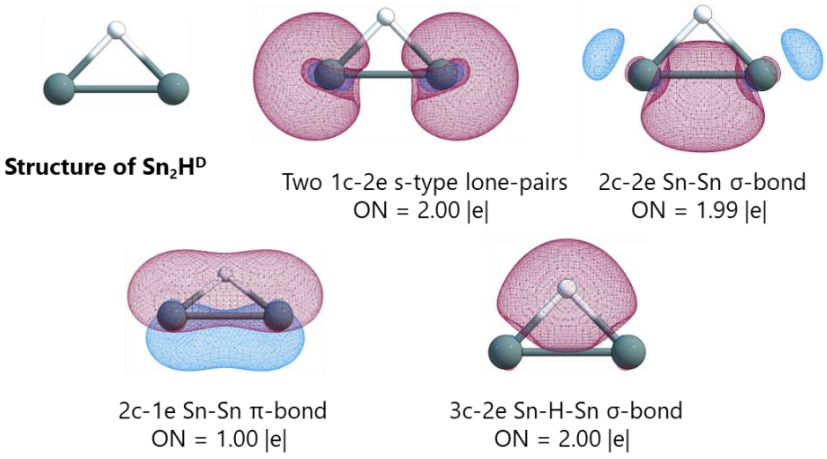


Figure S7-A. AdNDP analysis of Sn_2H

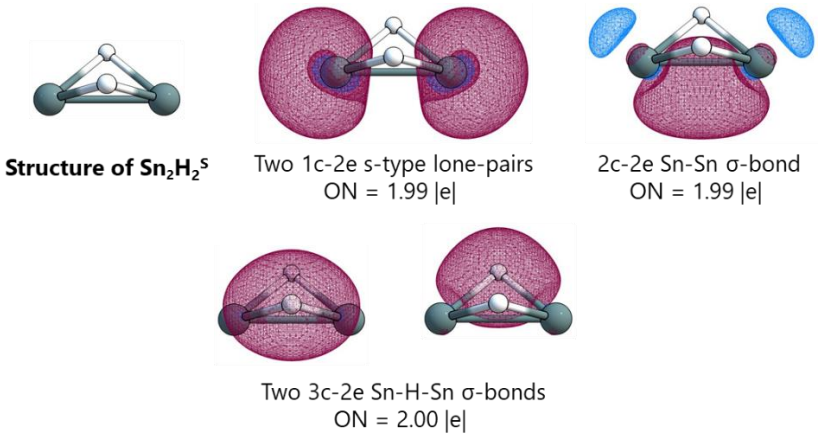


Figure S7-B. AdNDP analysis of Sn_2H_2

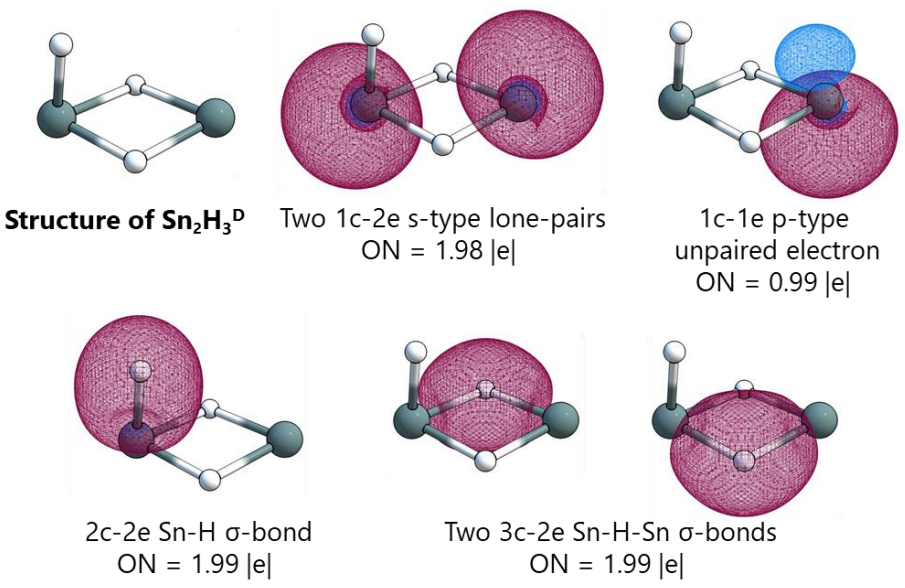


Figure S7-C. *AdNDP analysis of Sn_2H_3*

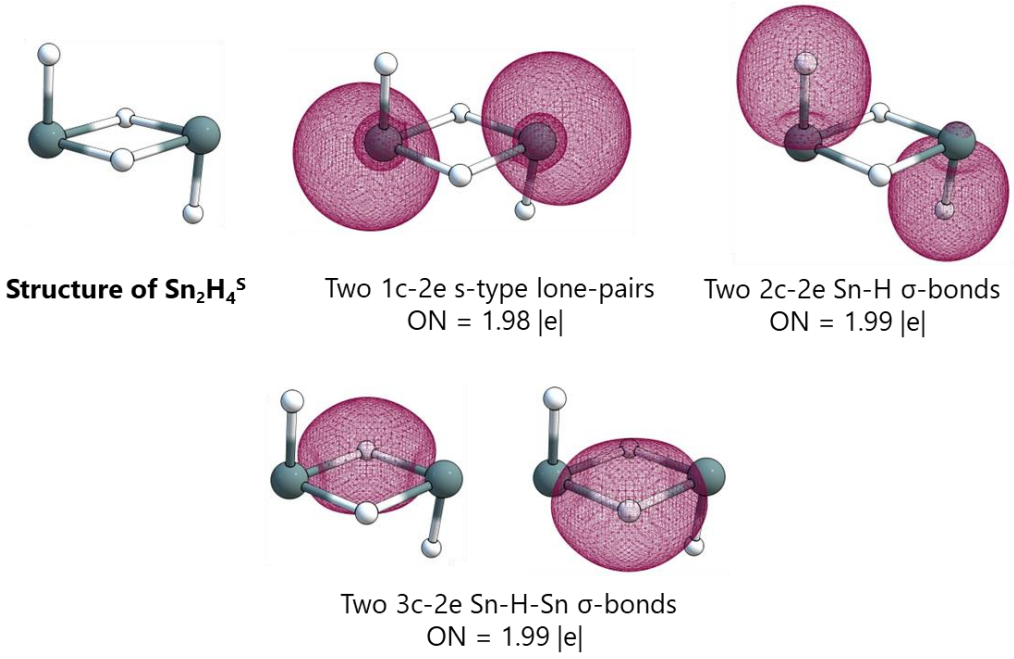


Figure S7-D. *AdNDP analysis of Sn_2H_4*

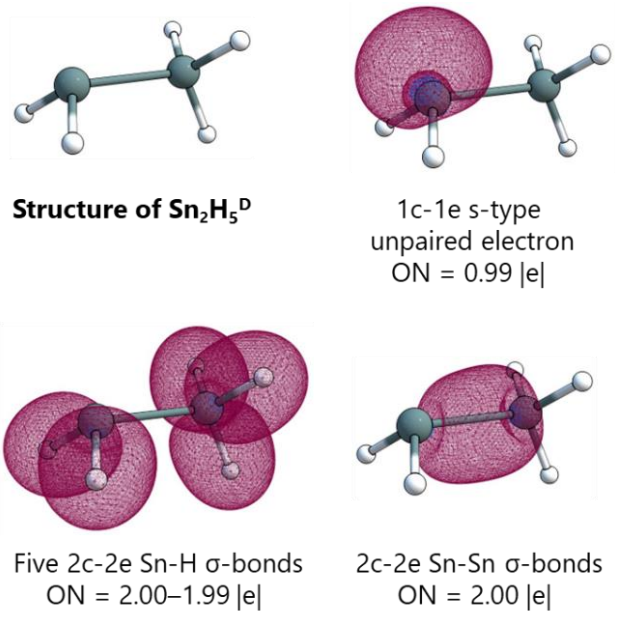


Figure S7-E. *AdNDP analysis of Sn_2H_5*

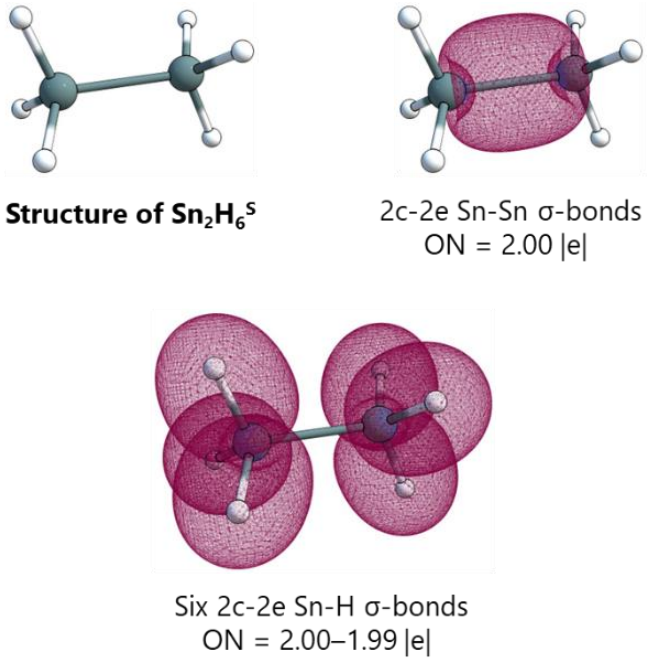


Figure S7-F. *AdNDP analysis of Sn_2H_6*

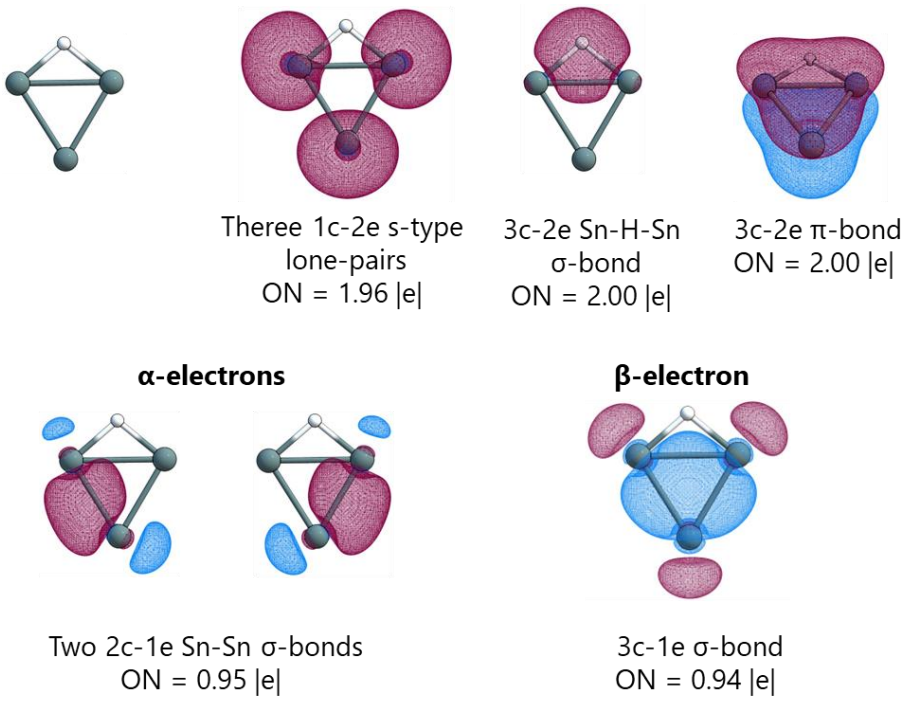


Figure S7-G. *AdNDP analysis of Sn_3H*

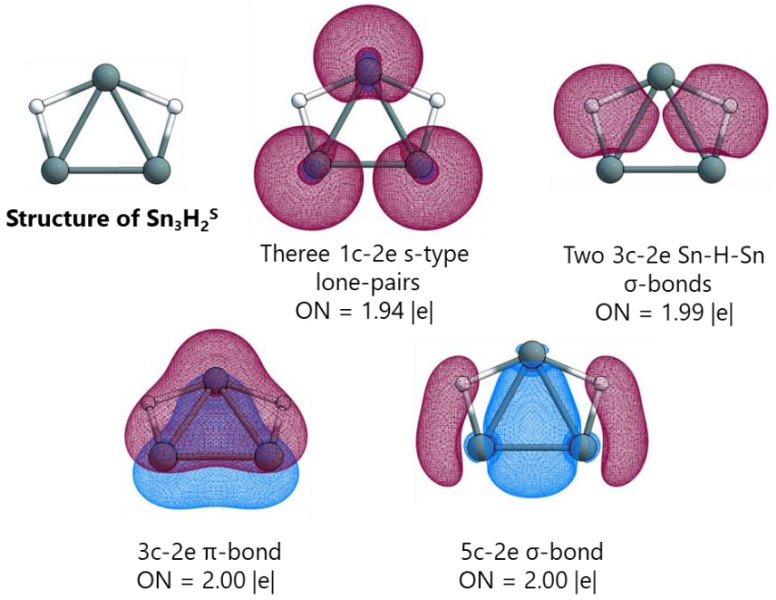


Figure S7-H. *AdNDP analysis of Sn_3H_2*

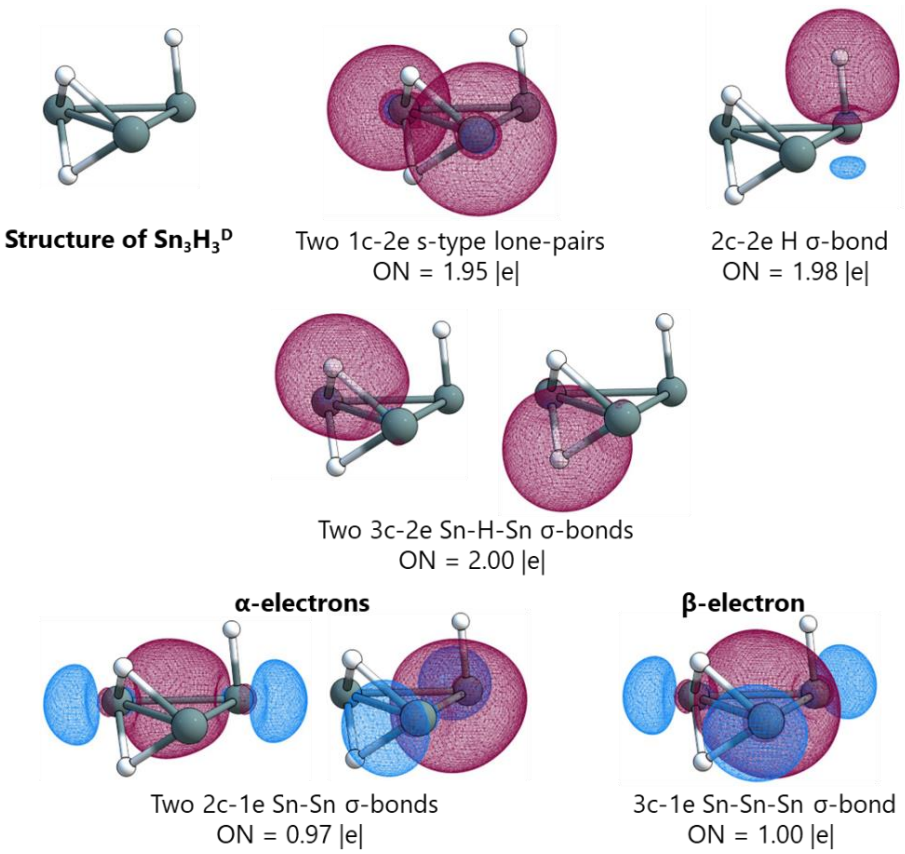


Figure S7-I. *AdNDP analysis of Sn_3H_3*

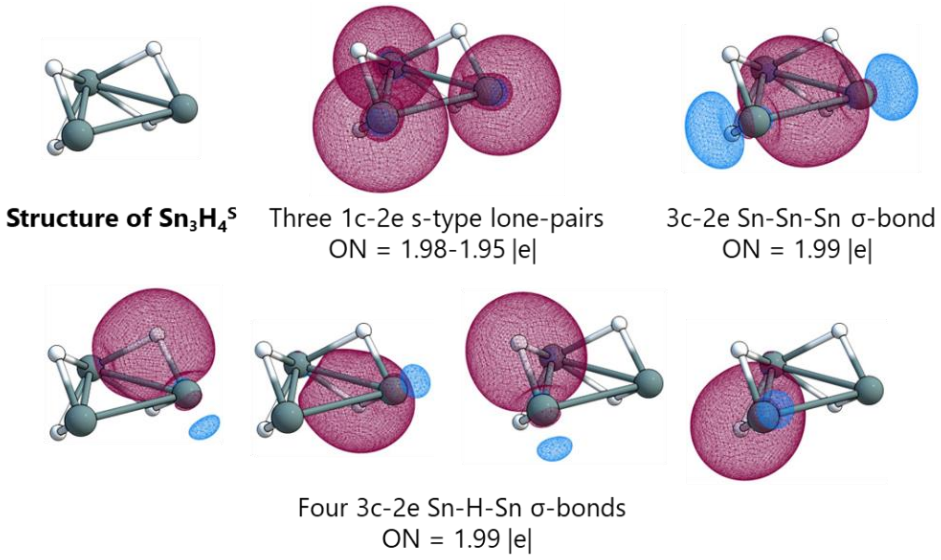


Figure S7-J. *AdNDP analysis of Sn_3H_4*

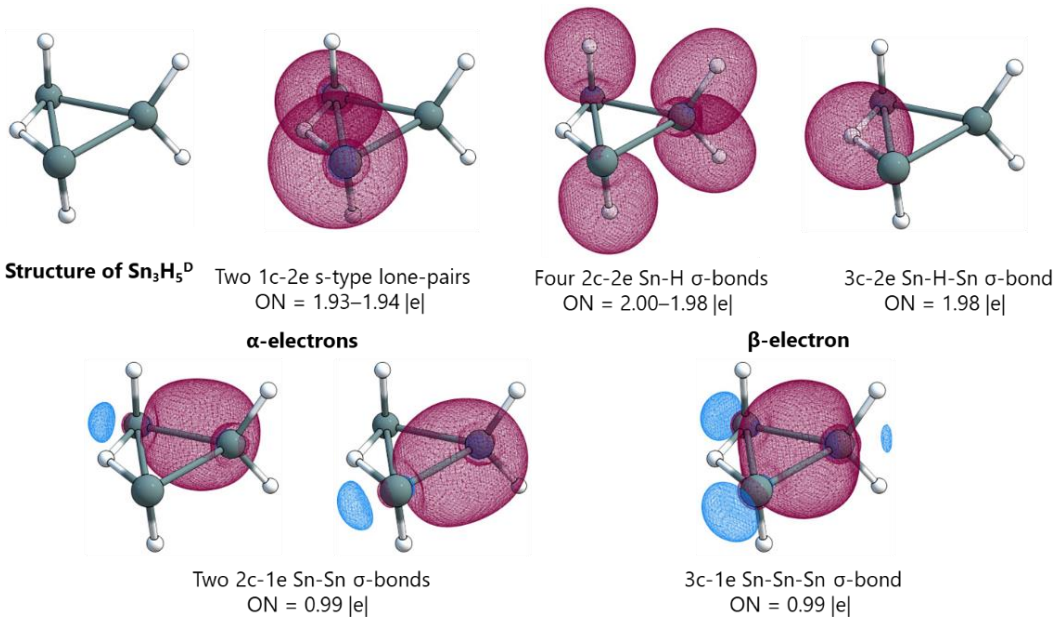


Figure S7-K. *AdNDP analysis of Sn_3H_5*

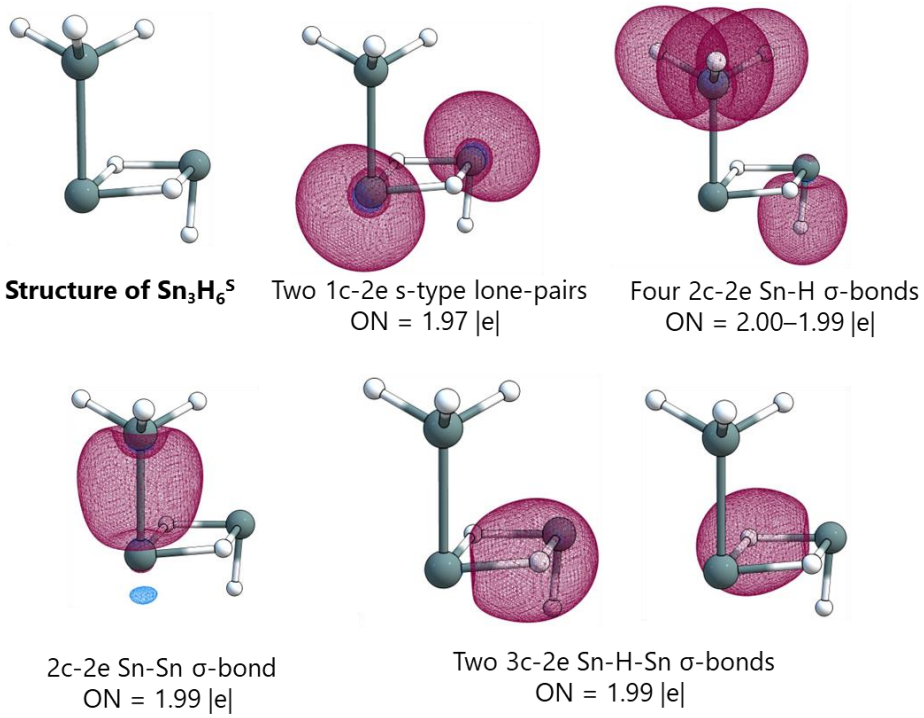
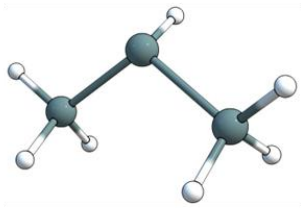
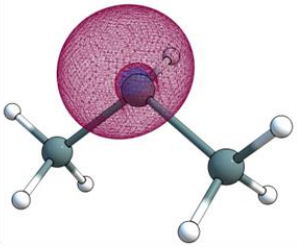


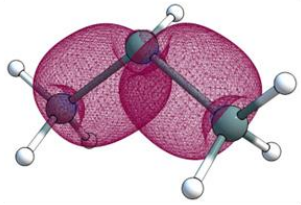
Figure S7-L. *AdNDP analysis of Sn_3H_6*



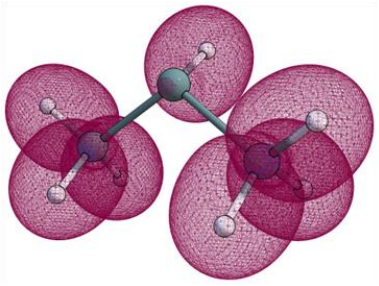
Structure of $Sn_3H_7^D$



1c-1e s-type unpaired electron
ON = 0.98 |e|

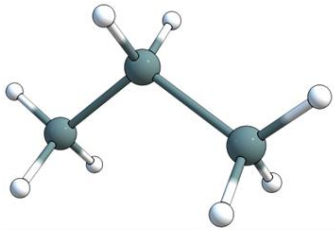


Two 2c-2e Sn-Sn σ-bonds
ON = 1.99 |e|

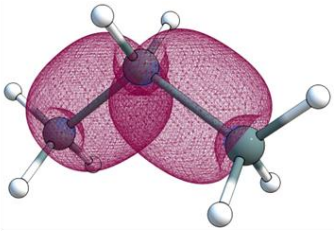


Seven 2c-2e Sn-H σ-bonds
ON = 1.99 |e|

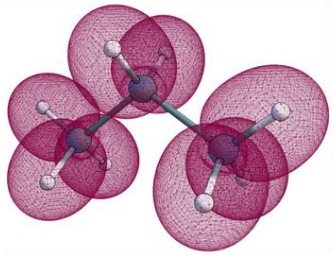
Figure S7-M. *AdNDP analysis of Sn_3H_7*



Structure of $Sn_3H_8^S$



Two 2c-2e Sn-Sn σ-bonds
ON = 1.99 |e|



Eight 2c-2e Sn-H σ-bonds
ON = 2.00–1.99 |e|

Figure S7-N*. AdNDP analysis of Sn_3H_8*

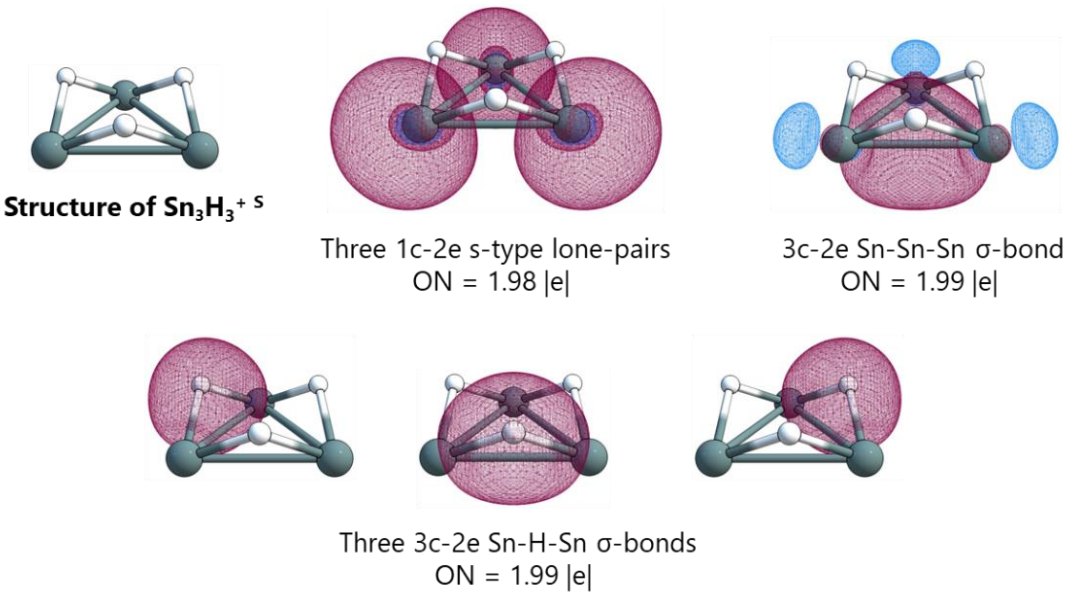


***Figure S7-O**. AdNDP analysis of $Sn_3H_3^+$*

S8. AdNDP analysis of Sn_2H_x and Sn_3H_y classic-like structures

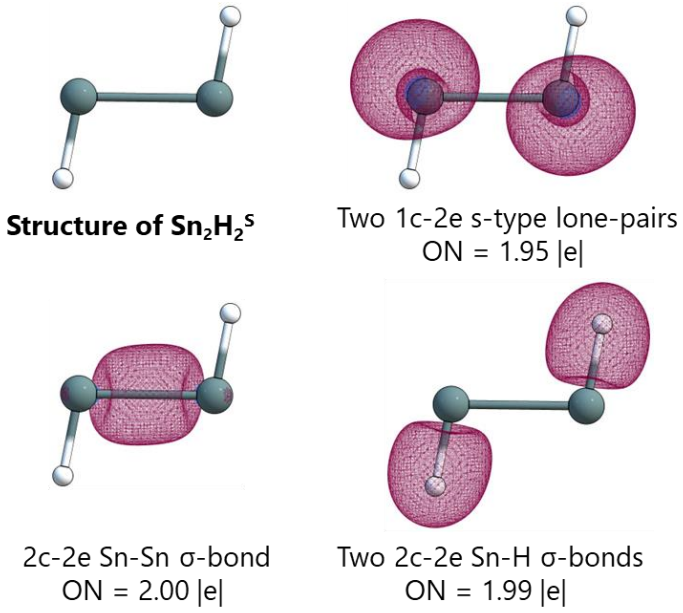


***Figure S8-A**. AdNDP analysis of classic-like Sn_2H_2*

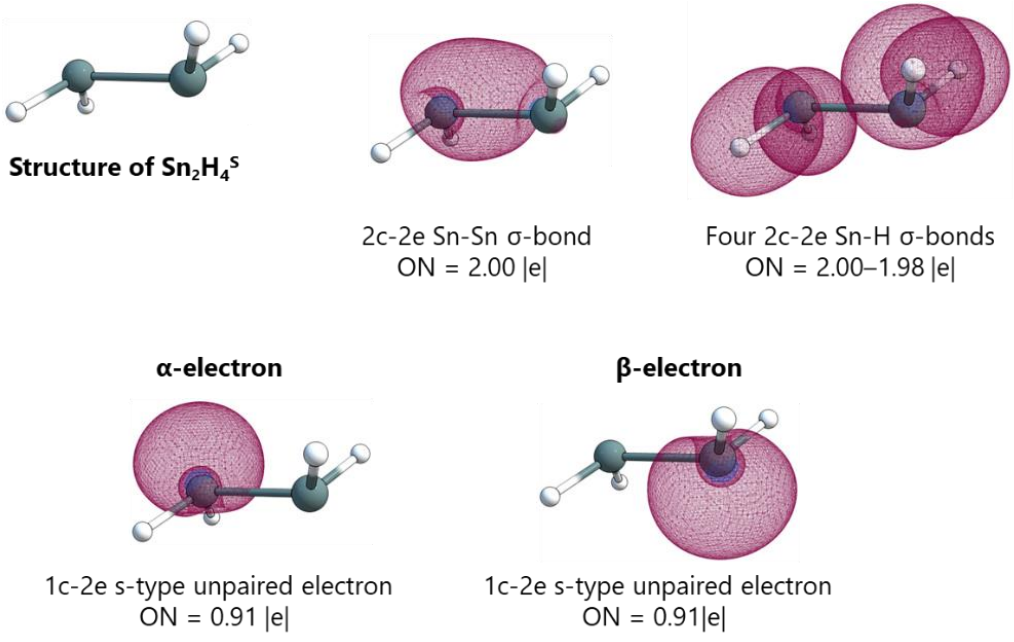


***Figure S8-B**. AdNDP analysis of classic-like Sn_2H_4*

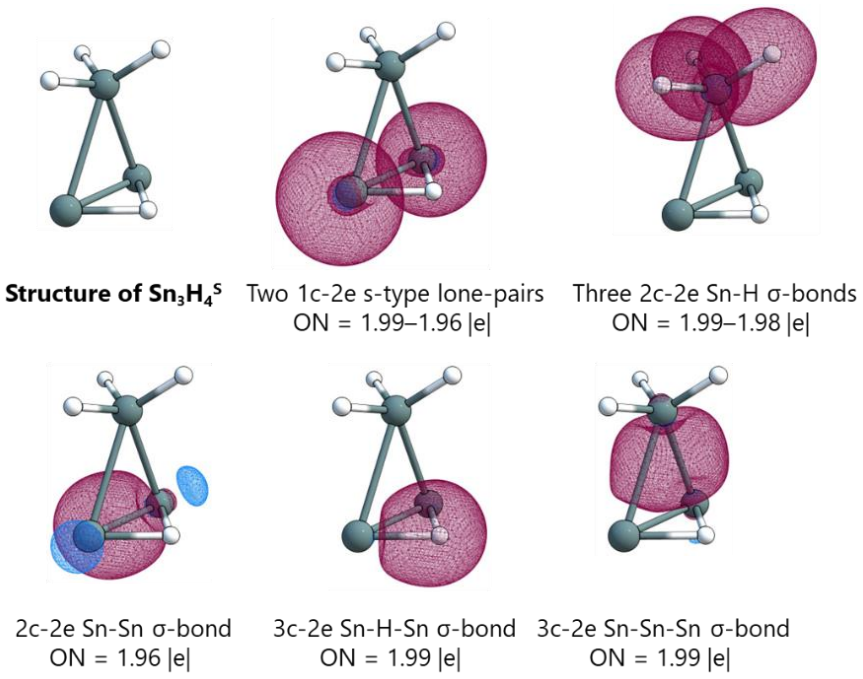


***Figure S8-C**. AdNDP analysis of classic-like Sn_3H_4*

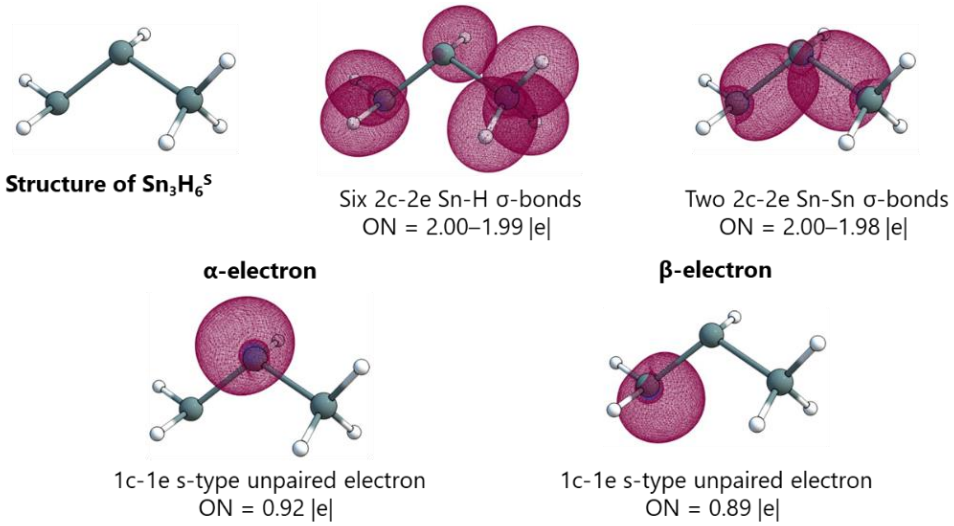


***Figure S8-D**. AdNDP analysis of classic-like Sn_3H_6*

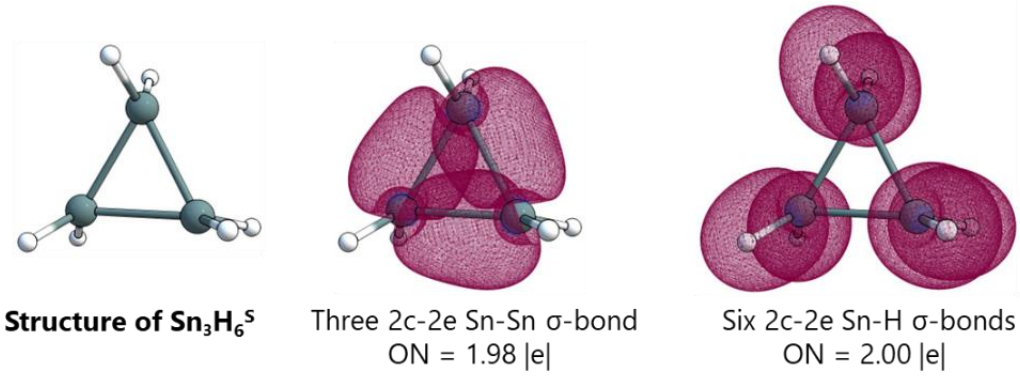


***Figure S8-E**. AdNDP analysis of cyclic Sn_3H_6*

S9. Coordinates (XYZ) of classic-like Sn_2H_x and Sn_3H_y structures

Table S9. *Coordinates (XYZ) of classic-like Sn2Hx and Sn3Hy structures*

Stoichiometry	XYZ coordinates			
Sn_2H_2	Sn	-0.052107602	0.253457424	-2.161585874
	Sn	0.305843612	-0.088544125	-4.987446994
	H	-0.868668658	1.082565992	-5.597212926
	H	1.124761157	-0.915389698	-1.552007619
Sn_2H_4	Sn	0.528420792	-1.038109843	0.601377775
	Sn	0.458779058	1.651223782	0.068519251
	H	-1.043806132	-1.737300185	0.791649764
	H	1.355792278	-1.420522061	2.073171000
	H	-0.369209987	2.033071807	-1.403085620
	H	2.030723990	2.350836499	-0.122431170
Sn_3H_4	Sn	-3.195421975	-0.714993330	-2.320060726
	Sn	-0.572124236	0.004624052	-3.526049169
	Sn	-1.480155297	0.297186907	-0.415219827
	H	-1.031696430	1.097304871	-4.772471808
	H	-1.921273021	-1.537000617	-0.970379118
	H	0.153364563	-1.385108927	-4.227393831
	H	0.700371967	0.798434152	-2.645934895
Aliphatic Sn_3H_6	Sn	-0.075224180	-0.404509482	-2.554570176
	Sn	-0.162600956	-0.038611426	-5.266461647
	Sn	1.815039631	1.277764306	-1.291786391
	H	-1.395350176	-0.901869692	-6.118878653
	H	1.310705578	-0.429468231	-6.083180642
	H	-1.618332059	-0.029158342	-1.857657580
	H	1.215392467	2.867284087	-1.035202599
	H	3.230664236	1.372061888	-2.261228216
	H	2.249233572	0.612675196	0.232396297
Cyclic Sn_3H_6	Sn	-2.046368070	-1.339217661	-1.498850461
	Sn	0.141085460	0.276043867	-2.344438292
	Sn	-0.215591098	-0.302394718	0.424113049
	H	0.037698416	1.957213715	-2.707789658
	H	1.621936613	-0.332952307	-2.981209078
	H	-0.953965024	0.762464752	1.559960106
	H	0.630727064	-1.527612013	1.291016215
	H	-1.996569787	-2.939172669	-2.136911363
	H	-3.583163575	-0.650713965	-1.864243519