

Supplemental Information for:

The Composition and Constitution of Compressed Strontium Polyhydrides

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Structure Searches – Technical Details

<u>Stoichiometry</u>	<u>Pressure (GPa)</u>	<u># of formula units</u>		<u>Stoichiometry</u>	<u>Pressure (GPa)</u>	<u># of formula units</u>
<u>SrH₂</u>	50	2		<u>SrH₅</u>	50	2
	50	3			100	2
	50	4			150	2
	100	2		<u>SrH₆</u>	50	2
	150	2			100	2
	150	3			150	2
<u>SrH₃</u>	50	2		<u>SrH₇</u>	50	2
	100	2			100	2
	100	3			150	2
	100	4		<u>SrH₈</u>	50	2
	150	2			100	2
	150	3			150	2
	150	4		<u>SrH₉</u>	50	2
<u>SrH₄</u>	50	2			100	2
	50	3			150	2
	50	4		<u>SrH₁₀</u>	100	2
	100	2			100	2
	100	3			150	2
	100	4		<u>SrH₁₁</u>	50	2
	150	2			100	2
	150	3			150	2
	150	4		<u>SrH₁₂</u>	50	2
					100	2
					150	2

Table T1. Details regarding the number of structure searches run on SrH_n systems.

SrH_n “Tie-Line” Plot

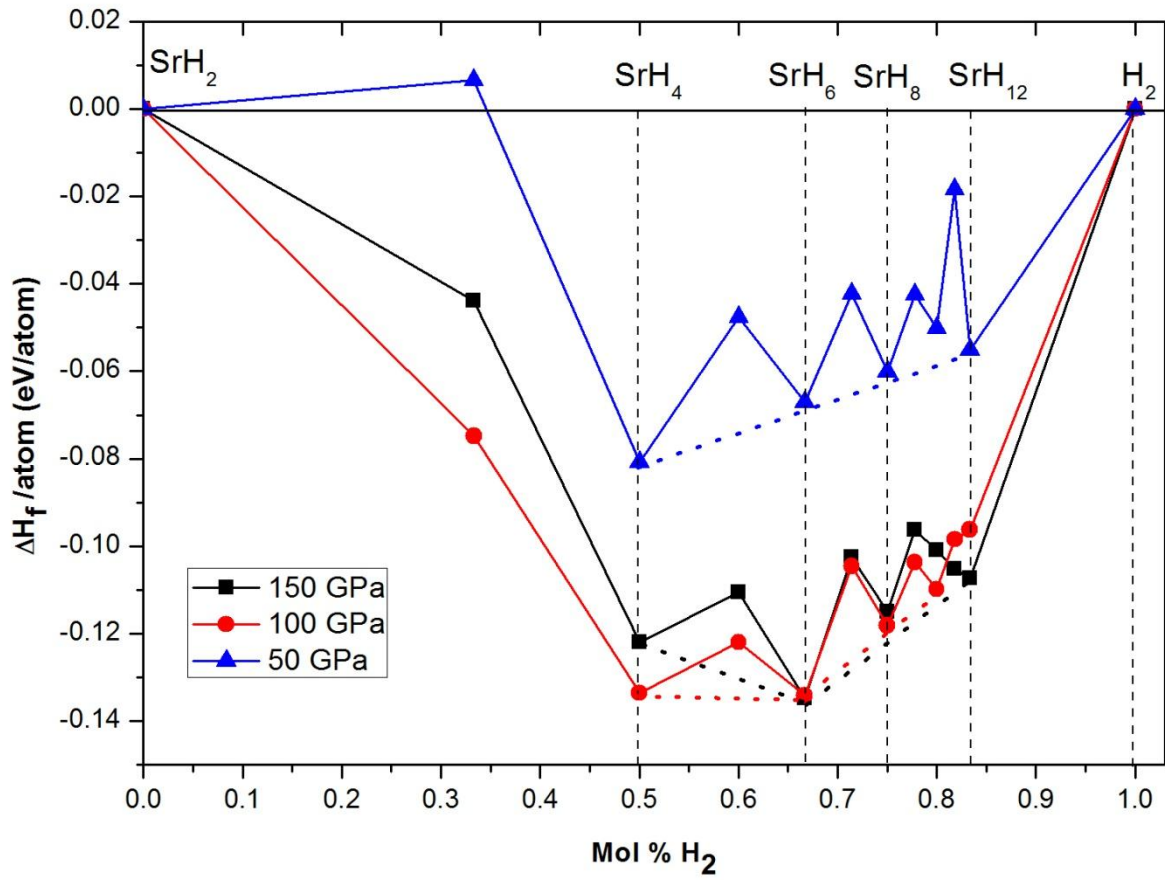


Figure S1. The enthalpy of formation, ΔH_f , for the reaction $SrH_2 + 1/2 (H_2)_{n-2} \rightarrow SrH_n$ versus the H_2 composition at varying pressures. The dashed blue, red and black lines represent the convex hulls at 50, 100, and 150 GPa, respectively. The enthalpy of SrH_2 and H_2 are computed for the most stable structures as described in the main text. The symbols represent points at which structure searches were explicitly run (see Table T1 for further search details).

Structural Parameters and Atomic Positions

<u>SrH₂ (Pnma)</u>			
a, b, c [Å]	5.633	4.098	7.097
α , β , γ	90.000	90.000	90.000
H (4c)	0.7487	0.2500	0.0834
H (4c)	0.0002	0.2500	0.7507
Sr (4c)	0.7502	0.2500	0.4167

Table T2. Calculated structural parameters of SrH₂ at 1 GPa.

<u>SrH₂ (P6₃/mmc)</u>			
a, b, c [Å]	3.608	3.608	4.519
α , β , γ	90.000	90.000	120.000
H (2c)	0.3333	0.6667	0.2500
H (2a)	0.0000	0.0000	0.0000
Sr (2d)	0.3333	0.6667	0.7500

Table T3. Calculated structural parameters of SrH₂ at 50 GPa.

<u>SrH₂ (P6/mmm)</u>			
a, b, c [Å]	3.148	3.148	2.665
α , β , γ	90.000	90.000	120.000
H (2d)	0.3333	0.6667	0.5000
Sr (a)	0.0000	0.0000	0.0000

Table T4. Calculated structural parameters of SrH₂ at 150 GPa.

<u>SrH₄ (I4/mmm)</u>			
a, b, c [Å]	3.304	3.304	5.650
α , β , γ	90.000	90.000	90.000
H (4d)	0.0000	0.5000	0.2500
H (4e)	0.0000	0.0000	0.5700
Sr (2a)	0.0000	0.0000	0.0000

Table T5. Calculated structural parameters of SrH₄ at 50 GPa.

<u>SrH₄ (Cmcm)</u>			
a, b, c [Å]	2.769	6.585	4.686
α , β , γ	90.000	90.000	112.810
H (8f)	0.0000	0.2106	0.8369
H (8f)	0.0000	0.6272	-0.0794
Sr (4c)	0.0000	0.0877	0.2500

Table T6. Calculated structural parameters of SrH₄ at 150 GPa.

<u>SrH₆ (C2/c)</u>			
a, b, c [Å]	3.066	10.549	3.641
α , β , γ	90.000	80.589	90.000
H (8f)	0.5980	0.4808	-0.0590
H (4e)	0.0000	0.5539	0.2500
H (4e)	0.0000	0.7370	0.2500
H (8f)	0.3823	0.3400	0.7162
Sr (4e)	0.0000	0.3622	0.2500

Table T7. Calculated structural parameters of SrH₆ at 100 GPa.

SrH₆ ($P\bar{3}$)			
a, b, c [Å]	5.370	5.370	3.011
α, β, γ	90.000	90.000	120.000
H (6g)	0.2784	-0.00886	0.5341
H (6g)	0.6310	0.6782	0.79984
H (6g)	-0.0613	0.3369	0.1455
Sr (1a)	0.0000	0.0000	0.0000
Sr (2d)	0.3333	0.6667	0.3325

Table T8. Calculated structural parameters of SrH₆ at 150 GPa.

SrH₆ ($R\bar{3}m$)			
a, b, c [Å]	5.397	5.397	2.947
α, β, γ	90.000	90.000	120.000
H (18f)	0.7138	0.0000	0.0000
Sr (3b)	0.0000	0.0000	0.5000

Table T9. Calculated structural parameters of SrH₆ at 150 GPa.

SrH₆ ($R\bar{3}m$)			
a, b, c [Å]	5.129	5.129	2.777
α, β, γ	90.000	90.000	120.000
H (18f)	0.7183	0.0000	0.0000
Sr (3b)	0.0000	0.0000	0.5000

Table T10. Calculated structural parameters of SrH₆ at 250 GPa.

SrH₈ ($C2/m$)			
a, b, c [Å]	6.240	3.580	2.915
α, β, γ	90.000	94.033	90.000
H (18f)	0.2602	0.2014	0.1209
	0.5387	0.0000	0.1145
	0.3323	0.0000	0.4942
Sr (3b)	0.0000	0.0000	0.5000

Table T11. Calculated structural parameters of SrH₈ at 100 GPa.

SrH₁₀ (Cm)			
a, b, c [Å]	7.621	3.812	6.579
α, β, γ	90.000	103.218	90.000
H (2a)	0.1513	0.0000	0.1922
H (2a)	0.6299	0.0000	0.3830
H (4b)	0.8189	0.2348	0.4345
H (4b)	0.7822	0.2777	-0.0147
H (2a)	0.1385	0.0000	0.2967
H (2a)	0.8535	0.0000	0.1643
H (2a)	0.0801	0.0000	0.7246
H (2a)	0.0241	0.0000	0.5712
H (4b)	0.4343	0.2339	0.4321
H (2a)	0.0529	0.0000	0.0055
H (2a)	0.5880	0.0000	0.8498
H (4b)	0.3938	0.2917	-0.0670
H (2a)	0.7660	0.0000	0.2695
H (2a)	0.6084	0.0000	0.6639
H (2a)	0.1224	0.0000	0.4816
H (2a)	0.6212	0.0000	0.5567
H (2a)	0.5042	0.0000	0.1443
Sr (2a)	0.2533	0.0000	0.7305

Table T12. Calculated structural parameters of SrH₁₀ at 50 GPa.

SrH₁₀ (C2/c)			
a, b, c [Å]	5.258	5.225	5.506
α, β, γ	90.000	80.804	90.000
H (8f)	0.8543	0.3364	0.7531
H (8f)	0.8459	0.3030	-0.0982
H (8f)	0.3509	0.4557	0.0552
H (8f)	0.0808	0.0783	0.6690
H (8f)	0.2003	0.4223	-0.0872
Sr (4e)	0.0000	0.3055	0.2500

Table T13. Calculated structural parameters of SrH₁₀ at 100 GPa.

SrH₁₀ (C2/c)			
a, b, c [Å]	5.049	5.024	5.189
α, β, γ	90.000	81.811	90.000
H (8f)	0.8681	0.1027	0.6259
H (8f)	0.8558	0.1667	0.8754
H (8f)	0.1159	0.4135	0.8823
H (8f)	0.5850	0.1786	0.1313
H (8f)	0.7247	0.4668	0.4680
Sr (4e)	0.0000	0.2417	0.2500

Table T14. Calculated structural parameters of SrH₁₀ at 150 GPa.

SrH₁₂ (C2/c)			
a, b, c [Å]	9.124	3.587	6.774
α, β, γ	90.000	71.199	90.000
H (8f)	-0.0742	0.2490	0.5928
H (8f)	-0.0967	0.2508	-0.0487
H (8f)	0.8292	0.2510	-0.0803
H (8f)	0.2812	0.2540	0.0258
H (8f)	0.2108	0.4806	0.7948
H (8f)	0.2873	0.4746	0.2094
Sr (4e)	0.0000	0.2496	0.2500

Table T15. Calculated structural parameters of SrH₁₂ at 50 GPa.

SrH₁₂ (C2/m)			
a, b, c [Å]	6.774	3.587	4.726
α, β, γ	111.026	90.000	117.906
H (4i)	0.7313	0.0000	0.3520
H (4i)	0.3950	0.0000	0.3075
H (4i)	0.5005	0.0000	0.1585
H (4i)	-0.0570	0.0000	0.0625
H (8j)	0.2453	0.2277	-0.0764
Sr (2c)	0.0000	0.0000	0.5000

Table T16. Calculated structural parameters of SrH₁₂ at 50 GPa.

SrH₁₂ (C2/m)			
a, b, c [Å]	7.159	3.081	4.597
α, β, γ	90.000	45.168	90.000
H (4i)	0.6490	0.0000	0.6509
H (8j)	0.0791	0.2578	-0.0514
H (4i)	0.6452	0.0000	0.0687
H (4i)	0.2680	0.0000	0.8634
H (4i)	0.2698	0.0000	0.5933
Sr (2c)	0.0000	0.0000	0.5000

Table T17. Calculated structural parameters of SrH₁₂ at 150 GPa.

SrH₄ Phonon Densities of States at 50 GPa and 150 GPa

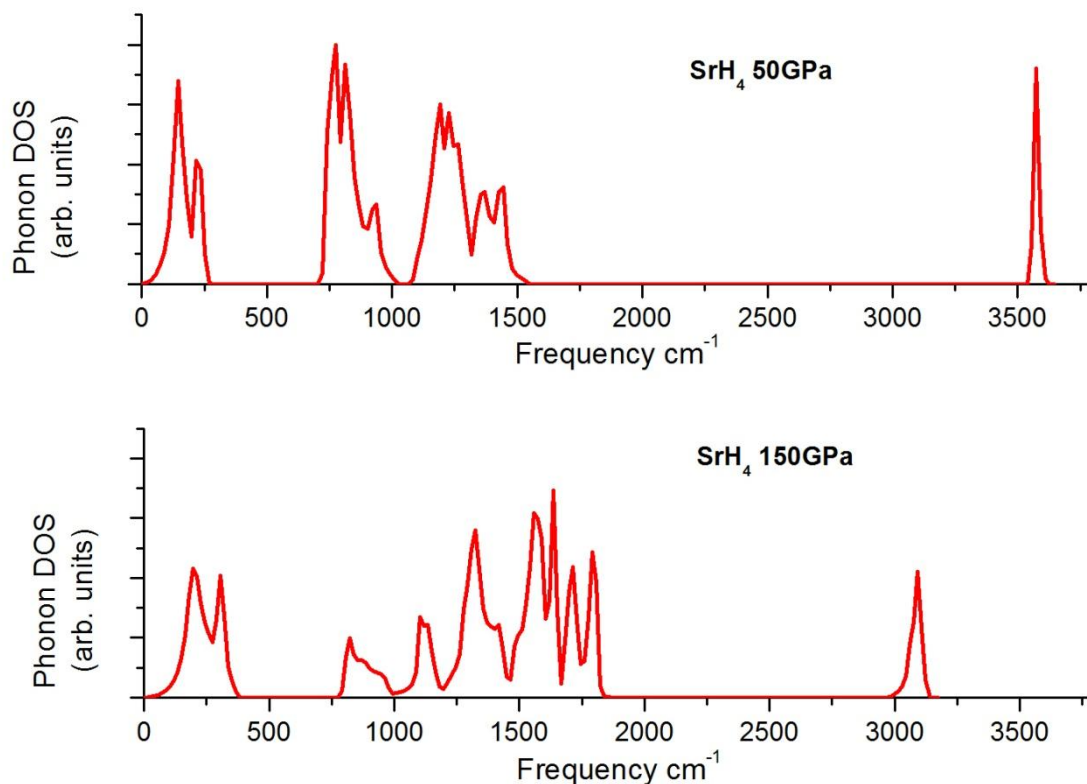


Figure S2. (top) The total phonon densities of states of the SrH₄ (*I4/mmm*) structure recovered at 50 GPa. (bottom) The total phonon densities of states of the lowest enthalpy SrH₄ structure (*Cmcm*) recovered at 150 GPa (also shown in the main text).

SrH₄ Projected Densities of States at 50 GPa and 150 GPa

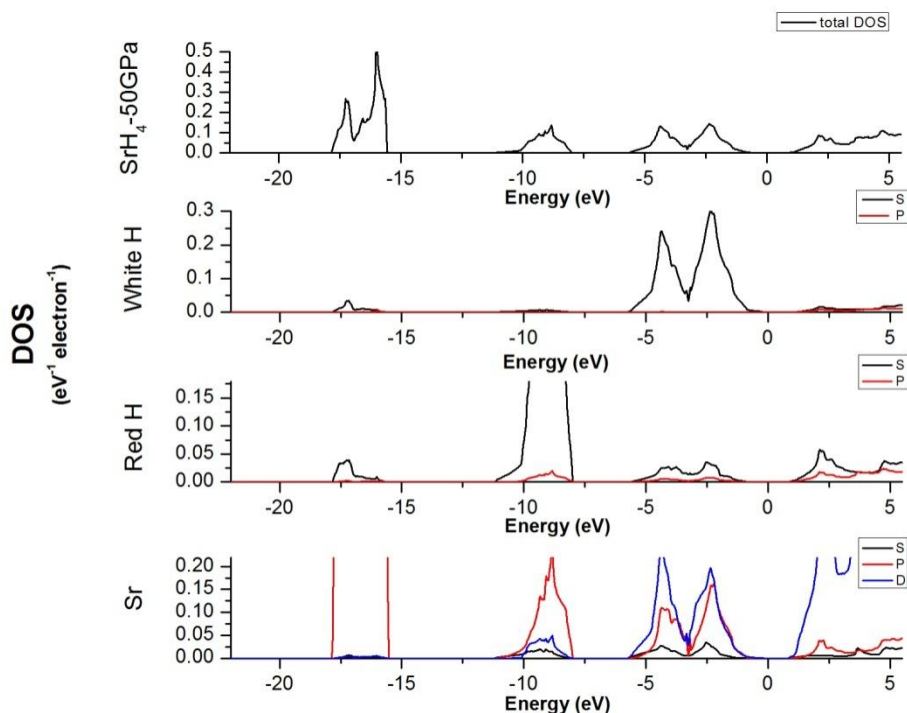


Figure S3. (top) The total electronic densities of states of the lowest enthalpy SrH₄ (*I4/mmm*) structure recovered at 50 GPa. (below) Below are shown the projected densities of states onto the hydrogen and Sr atoms as labeled in the main text. The Fermi energy is set to zero.

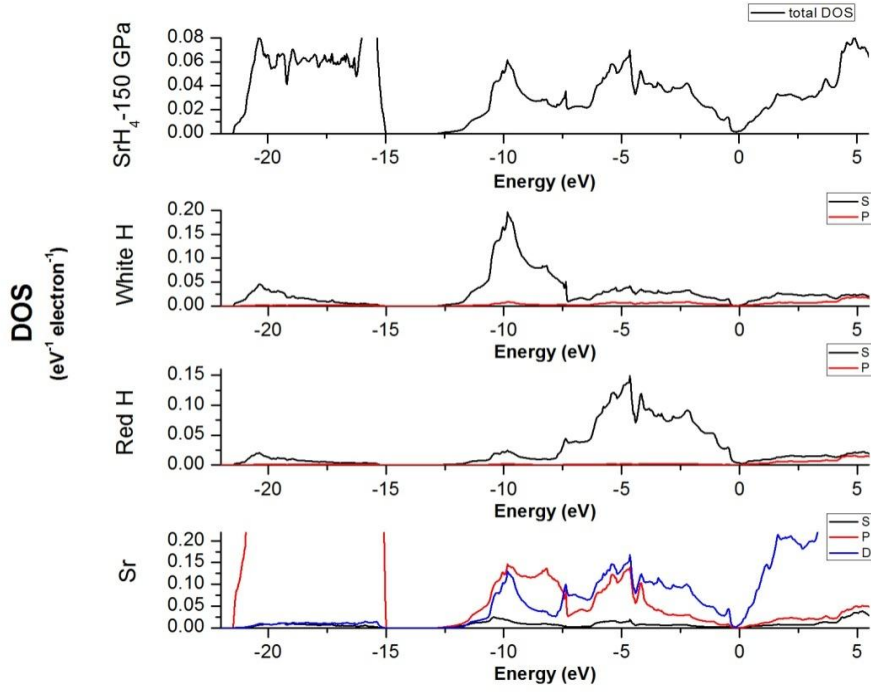


Figure S4. (top) The total electronic densities of states of the lowest enthalpy SrH_4 ($Cmcm$) structure recovered at 150 GPa. (below) Below are shown the projected densities of states onto the hydrogen and Sr atoms as labeled in the main text. The Fermi energy is set to zero.

SrH_6 Phonon Densities of States at 100, 150, and 250 GPa

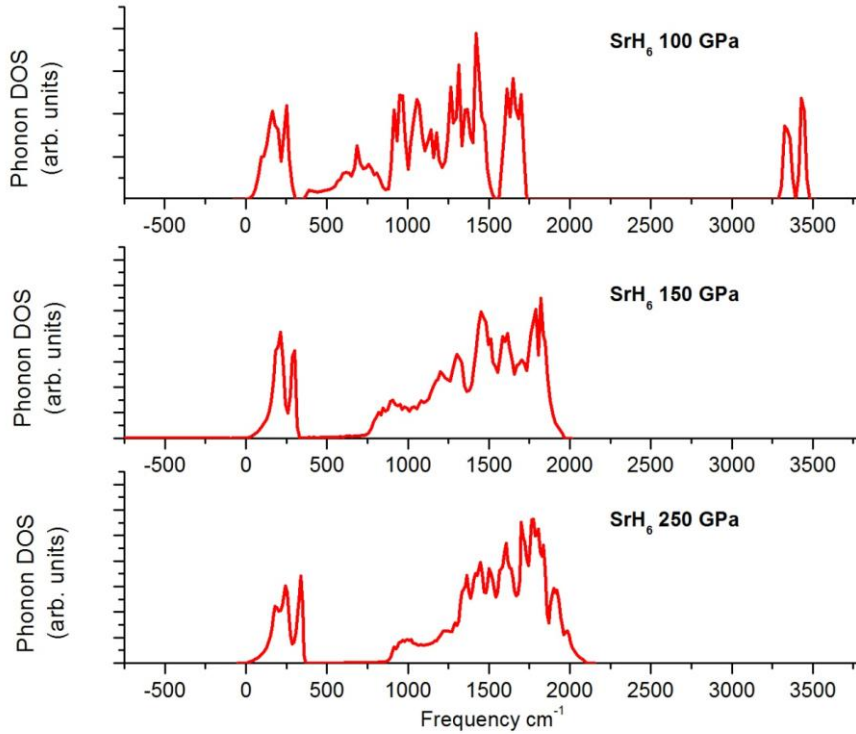


Figure S5. (top) The total phonon densities of states of the SrH_6 ($C2/c$) structure recovered at 100 GPa. (middle) The total phonon densities of states of the SrH_6 ($R\bar{3}m$) structure recovered at 150 GPa. The negative axis is given to show the extent of imaginary frequencies found for the ($R\bar{3}m$) SrH_6 structure at 150 GPa. In the other two plots all frequencies were real. (bottom) The total phonon densities of states of the SrH_6 ($R\bar{3}m$) structure at 250 GPa.

SrH₆ Projected Densities of States at 100 GPa

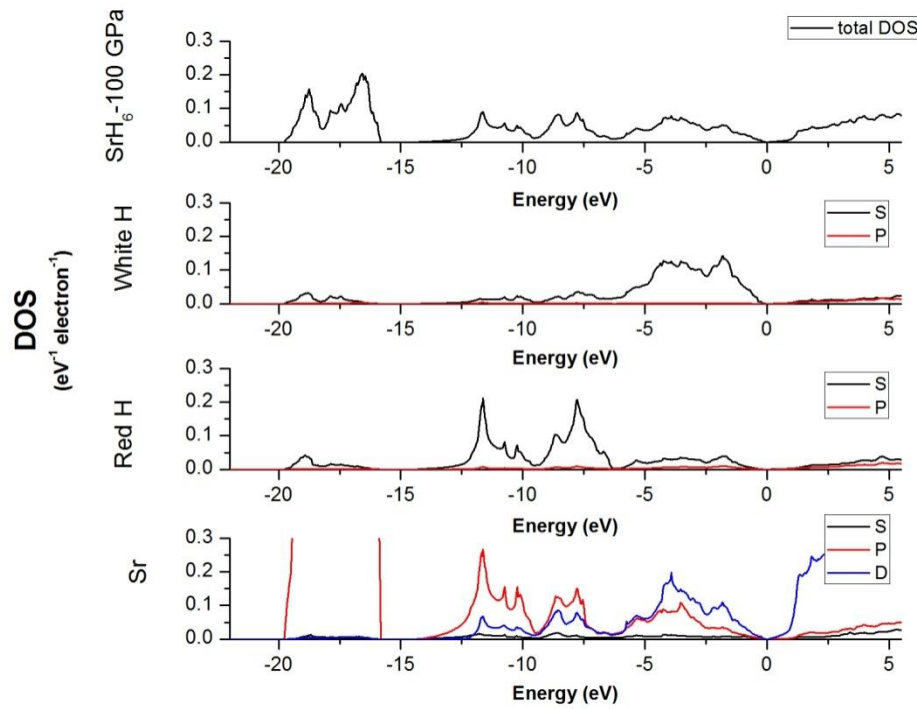


Figure S6. (top) The total electronic densities of states of the lowest enthalpy SrH₆ (*C2/c*) structure recovered at 100 GPa. (below) Below are shown the projected densities of states onto the hydrogen and Sr atoms as labeled in the main text. The Fermi energy is set to zero.

SrH₆ Structure and Total Electronic Densities of States at 150 and 250 GPa

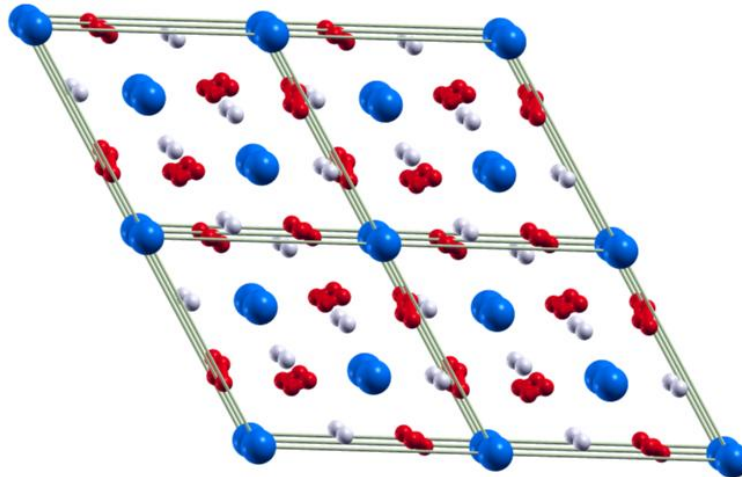


Figure S7. The extended structure of the lowest enthalpy phase ($P\bar{3}$) found upon following the largest soft phonon mode in SrH₆ ($R\bar{3}m$) at 150 GPa.

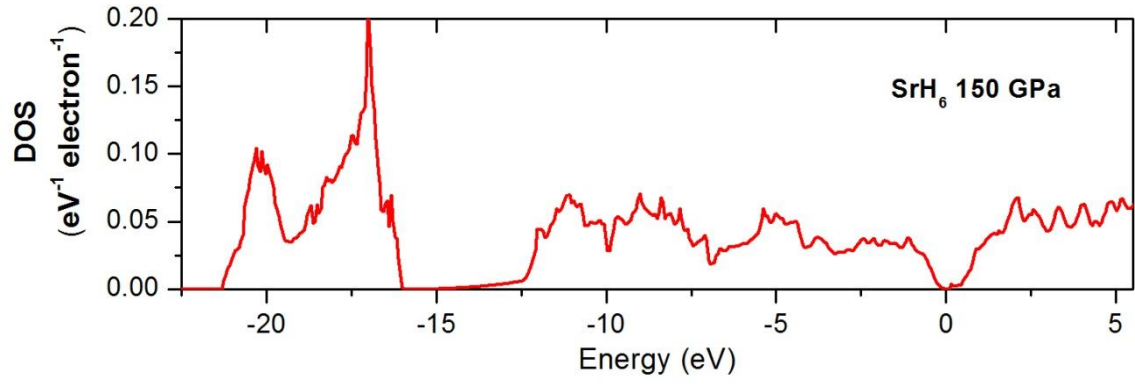


Figure S8. The total electronic densities of states of the lowest enthalpy SrH_6 ($P\bar{3}$) structure recovered by following the largest soft phonon mode in SrH_6 ($R\bar{3}m$) at 150 GPa. The Fermi energy is set to zero.

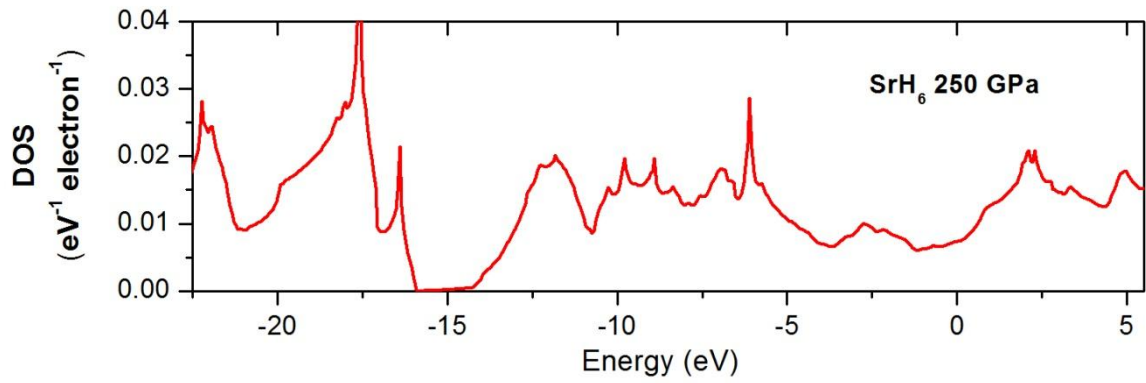


Figure S9. The total electronic densities of states of the lowest enthalpy SrH_6 ($R\bar{3}m$) structure recovered at 250 GPa. The Fermi energy is set to zero.

SrH₁₀ Structure and Total Electronic Densities of States at 100 GPa

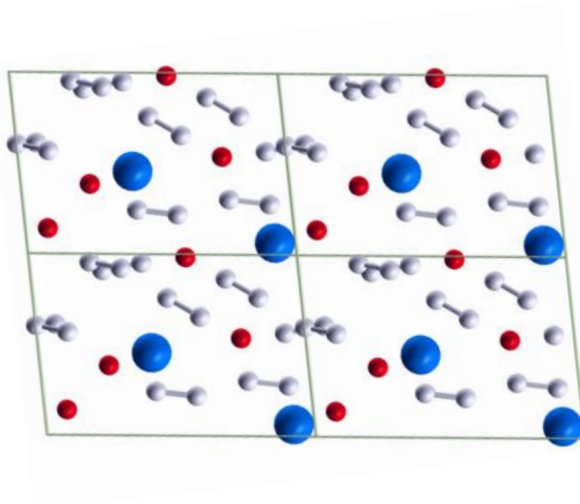


Figure S10. The extended structure of the lowest enthalpy SrH₁₀ structure recovered at 100 GPa (*Cm*). Note that the color scheme used to color the hydrogen sublattice is reversed from that used in the main text: hydridic hydrogen atoms are colored red and hydrogen atoms belonging to a dihydrogen motif are colored white.

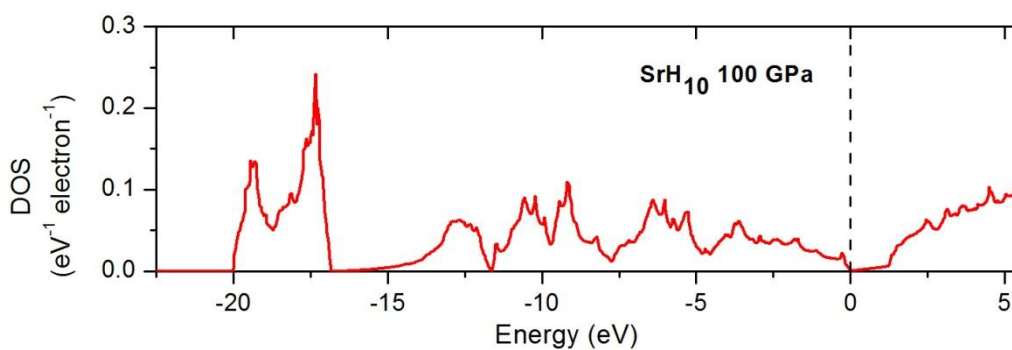


Figure S11. The total electronic densities of states of the lowest enthalpy SrH₁₀ (*Cm*) structure recovered at 100 GPa. The Fermi energy is set to zero.

SrH₁₀ Structure and Total Electronic Densities of States at 150 GPa

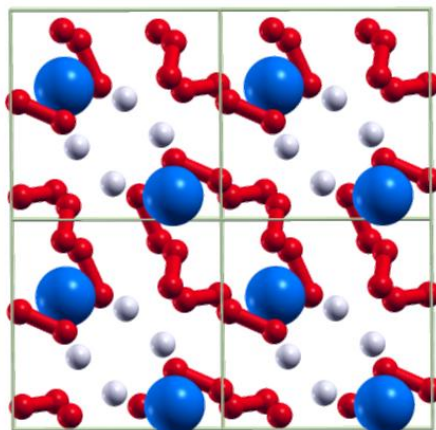


Figure S12. The extended structure of the lowest enthalpy SrH₁₀ structure at 150 GPa (*C2/c*).

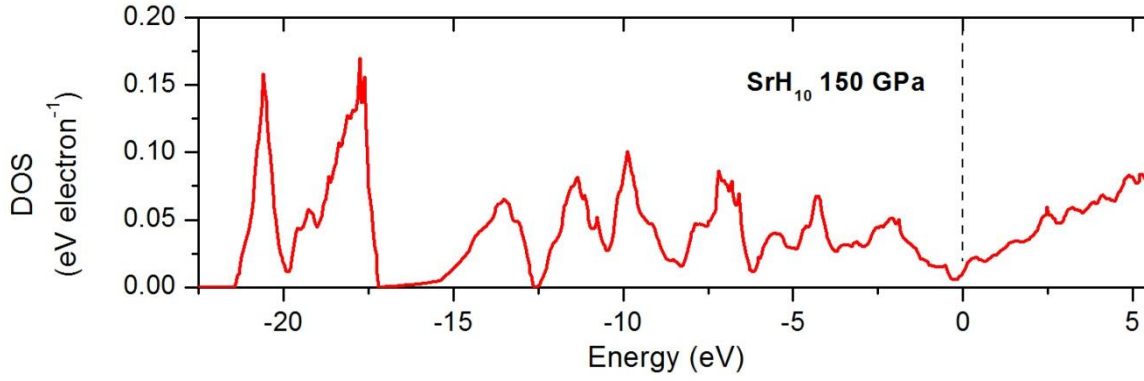


Figure S13. The total electronic densities of states for the lowest enthalpy SrH_{10} ($C2/c$) structure recovered at 150 GPa. Note: This structure does not lie on the convex hull at 150 GPa.

SrH_{12} Structure and Total Electronic Densities of States at 50 GPa

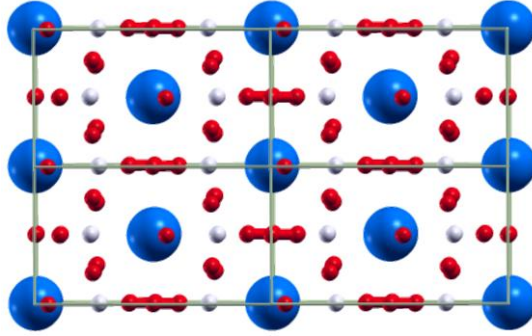


Figure S14. The extended structure of the isoenthalpic lowest energy structure of SrH_{12} recovered at 50 GPa ($C2/m$).

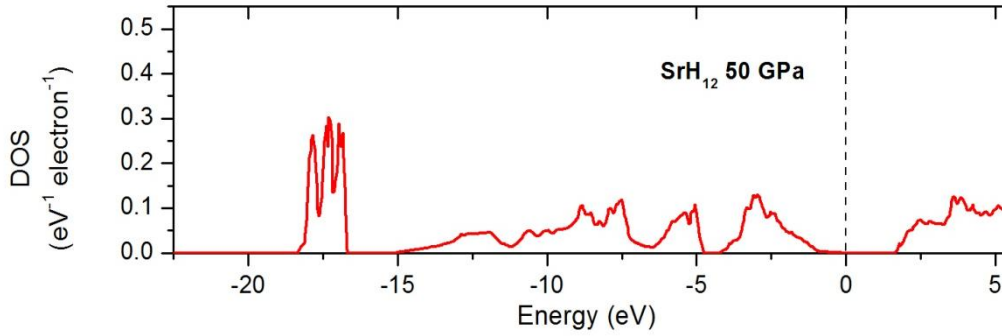


Figure S15. The total electronic densities of states of the other isoenthalpic lowest enthalpy structure of SrH_{12} recovered at 50 GPa ($C2/m$). The Fermi energy is set to zero.

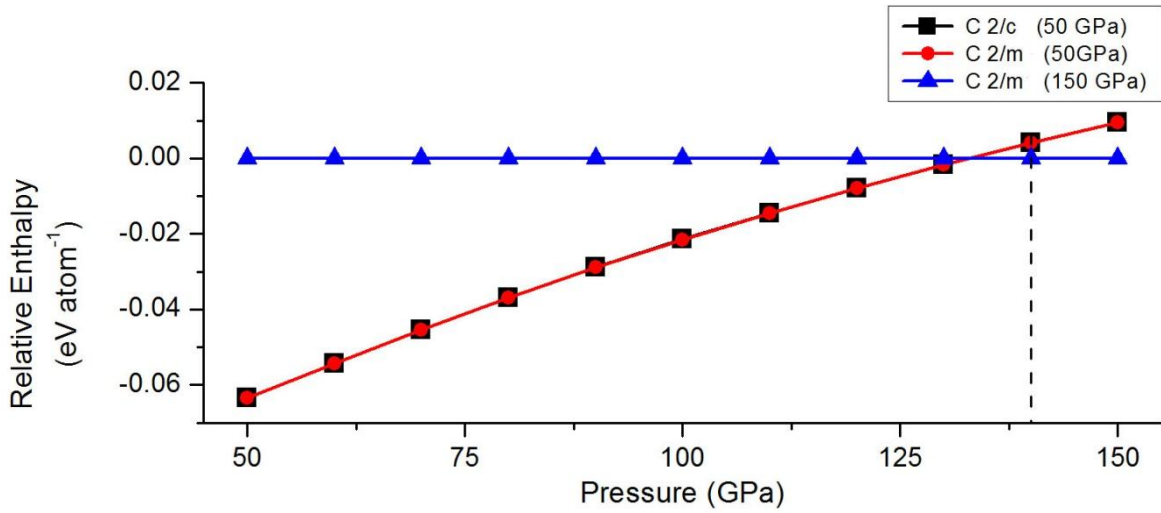


Figure S16. Relative enthalpy difference between isoenthalpic low pressure phases of SrH_{12} and the high pressure phase of SrH_{12} across the pressure range explored.

SrH_{12} Projected Densities of States at 150 GPa

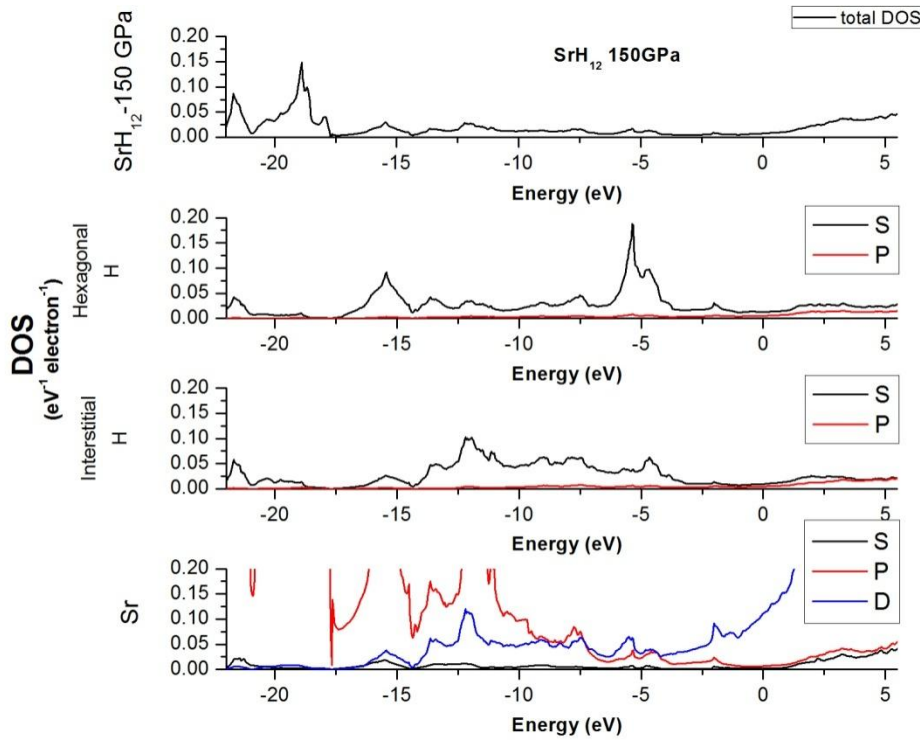


Figure S17. (top) The total electronic densities of states of the lowest enthalpy SrH_{12} ($C2/m$) structure recovered at 150 GPa. (below) Below are shown the projected densities of states onto the hydrogen and Sr atoms. The Fermi energy is set to zero.

HSE06 calculations on SrH₄ and SrH₆ at 150 GPa

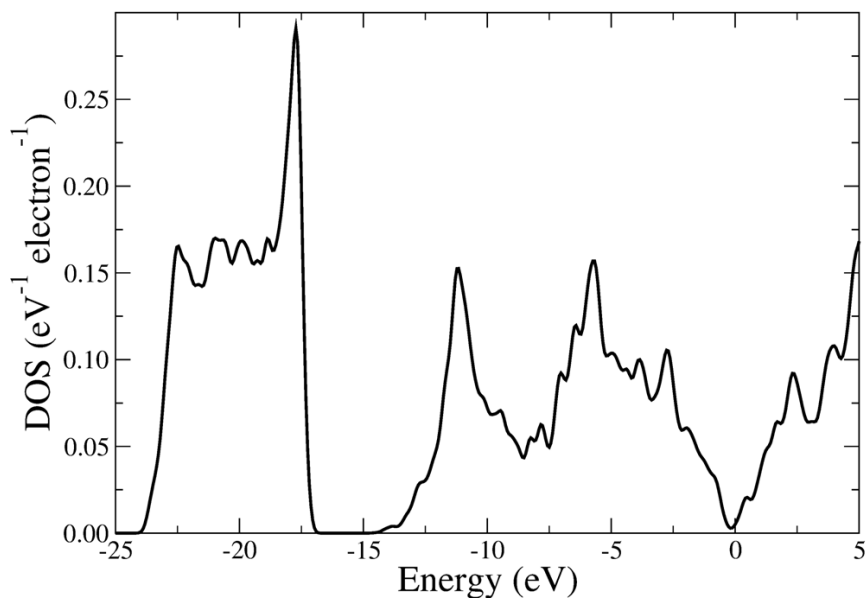


Figure S18. The total electronic densities of states of the lowest enthalpy SrH₄ (*Cmcm*) structure recovered at 150 GPa, computed with the hybrid HSE06 functional. The geometry was left in the PBE-optimized geometry. The Fermi energy is set to zero.

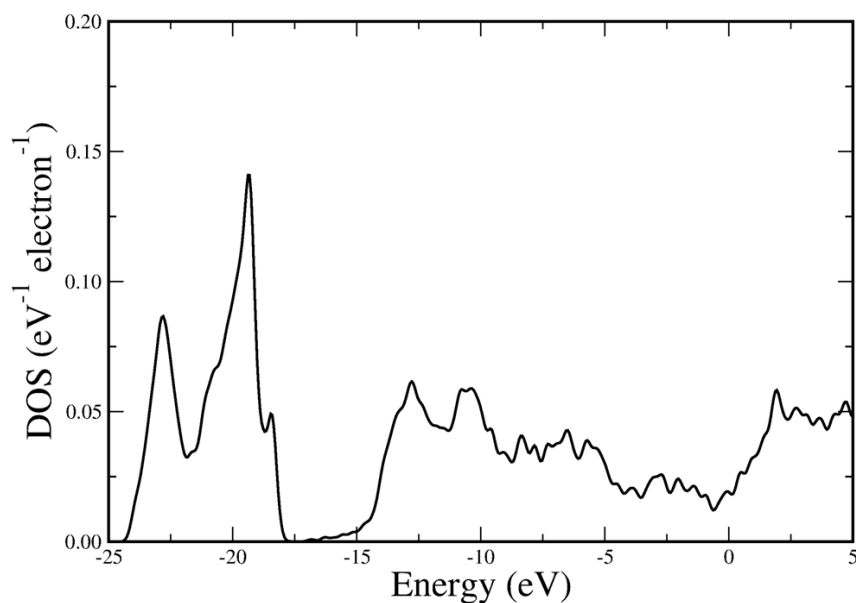


Figure S19. The total electronic densities of states of the lowest enthalpy SrH₆ (*R* $\bar{3}$ *m*) structure recovered at 150 GPa, computed with the hybrid HSE06 functional. The geometry was left in the PBE-optimized geometry. The Fermi energy is set to zero.