Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting information for

High-T_c superconductivity of polyhydride Rb_2MgH_{18} with a layered hydrogen structure at high pressure

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Table S1 Proposed possible reactions for synthesizing the Rb_2MgH_{18} phase and corresponding formation enthalpies at 250 GPa.

Reaction	Formation enthalpy (meV/atom)
$MgH_2+2RbH+7H_2 \rightarrow Rb_2MgH_{18}$	-53.2
$MgH_4+2RbH+6H_2 \rightarrow Rb_2MgH_{18}$	-34.8
$MgH_{12}+2RbH+2H_2 \rightarrow Rb_2MgH_{18}$	-21.9
$MgH_{16}+2RbH \rightarrow Rb_2MgH_{18}$	-19.8

Table S2 Predicted structures of Rb_2MgH_{18} at pressure of 250 GPa.

	Lattice parameter	Atom coordinates
*P*1	a = 2.611 Å b = 2.671 Å c = 9.574 Å α = 97.077° β = 96.272° γ = 61.057°	Rb(1a) 0.308999, 0.482208, 0.851413 Rb(1a) 0.376345, 0.574038, 0.131486 Mg(1a) 0.468237, 0.728198, 0.526023 H(1a) 0.407778, 0.658143, 0.345924 H(1a) 0.625895, 0.282581, 0.361020 H(1a) 0.875123, 0.695917, 0.283366 H(1a) 0.083008, 0.267492, 0.275114 H(1a) 0.242859, 0.025964, 0.337723 H(1a) 0.718381, 0.000533, 0.255991 H(1a) 0.965228, 0.210737, 0.991216 H(1a) 0.344581, 0.026805, 0.991495 H(1a) 0.723617, 0.842978, 0.991605 H(1a) 0.818594, 0.088547, 0.569746 H(1a) 0.400448, 0.323413, 0.639217 H(1a) 0.054104, 0.518181, 0.646444 H(1a) 0.882821, 0.128969, 0.717299 H(1a) 0.219383, 0.958407, 0.703857 H(1a) 0.654433, 0.738941, 0.701409 H(1a) 0.961393, 0.643178, 0.410106 H(1a) 0.125010, 0.350163, 0.457064 H(1a) 0.743661, 0.069940, 0.427942
*P*6/*mmm*	a = 5.093 Å c = 5.138 Å α = 90° β = 90° γ = 120°	Rb(3g) 0.50000, 0.00000, 0.50000 Rb(1a) 0.00000, 0.00000, 0.00000 Mg(2c) 0.66667, 0.33333, 0.00000 H(12o) 0.10233, 0.20465, 0.35081 H(12o) 0.60208, 0.80104, 0.20482 H(12n) 0.61680, 0.61680, 0.86765

Table S3 H-H bond lengths and volumes of unit cell of *P6/mmm* Rb_2MgH_{18} at 235 GPa calculated using PBE[1] and SCAN[2] functionals.

	PBE	**SCAN**
H1-H1	0.91 Å	0.89 Å
H2-H3	1.05 Å	1.04 Å
H1-H2	1.15 Å	1.17 Å
H3-H3	1.20 Å	1.21 Å
Volume	117.89 $Å^3$	118.0 $Å^3$

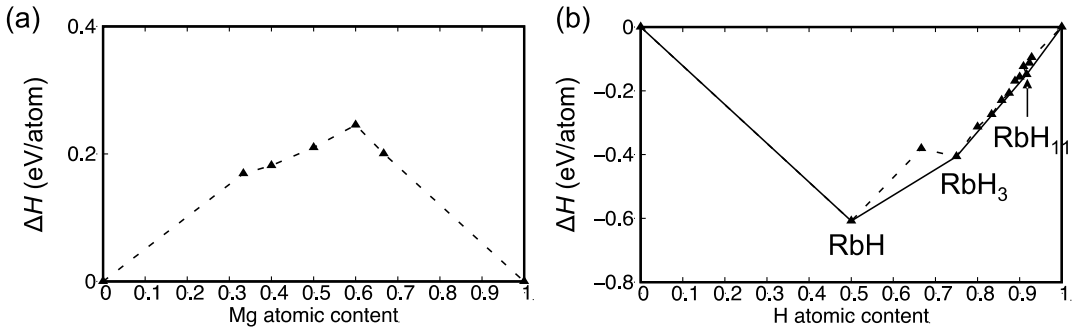

Fig. S1 Calculated convex hulls of (a) Rb-Mg and (b) Rb-H compounds at the pressure of 250 GPa.

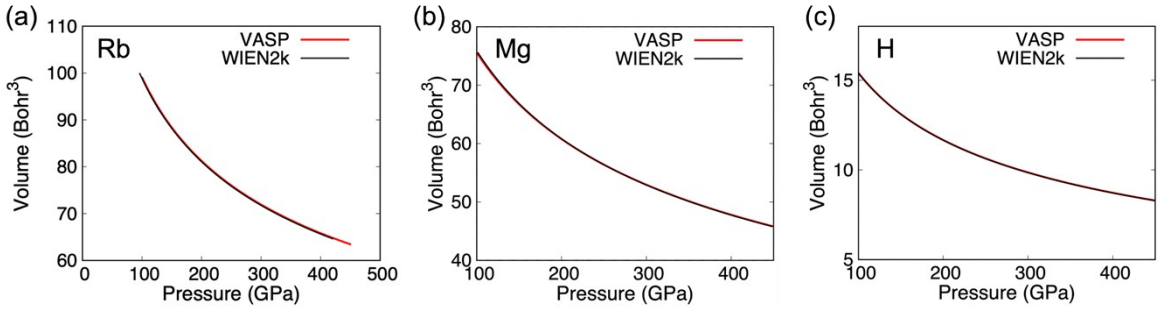

Fig. S2 Calculated equations of states for stable elemental phases of (a) Rb, (b) Mg, and (c) H with unit cells under high pressure using VASP and WIEN2K software.

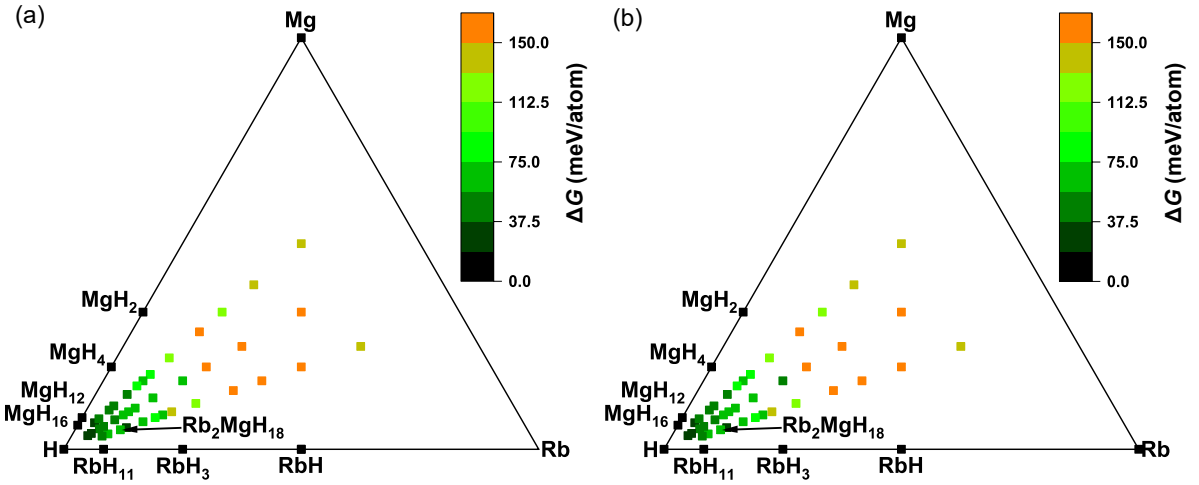

Fig. S3 Phase diagrams of $Rb_xMg_yH_z$ compounds relative to Rb, Mg, and H at (a) 180 K and 250 GPa, and (b) 280 K and 250 GPa. Black solid squares represent the stable phases, and the squares with the other colors mapped to the Gibbs free energies (meV/atom) above the convex hull indicate the metastable phases.

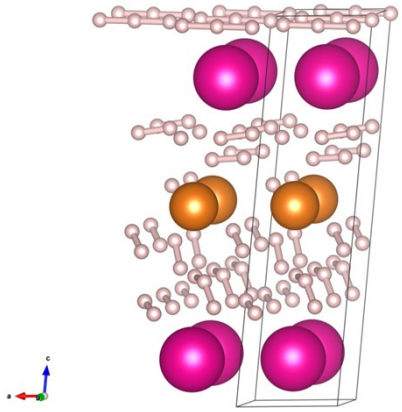

Fig. S4 Crystal structure of stable *P*1 phase of Rb_2MgH_{18}. The solid lines indicate the crystal lattice. The magenta, orange and light pink spheres represent the Rb, Mg and H, respectively.

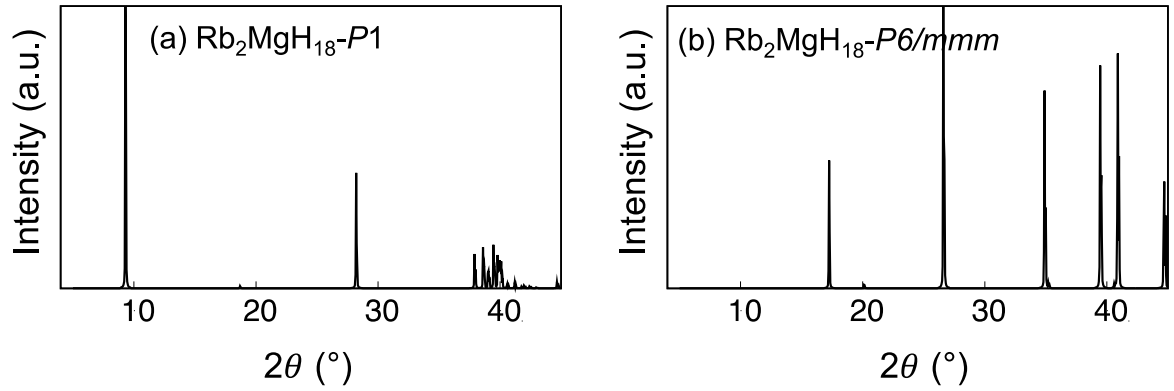

Fig. S5 Simulated X-ray diffraction patterns for (a) *P*1 and (b) *P*6/*mmm* phases of Rb_2MgH_{18} at 250 GPa.

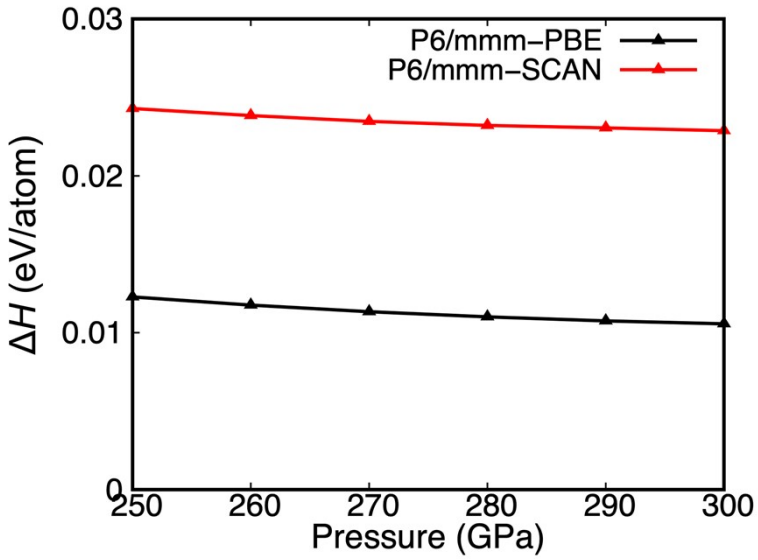

Fig. S6 Calculated enthalpies of *P*6/*mmm* structure with respect to *P*1 structure of Rb_2MgH_{18} using PBE[1] and SCAN[2] functionals.

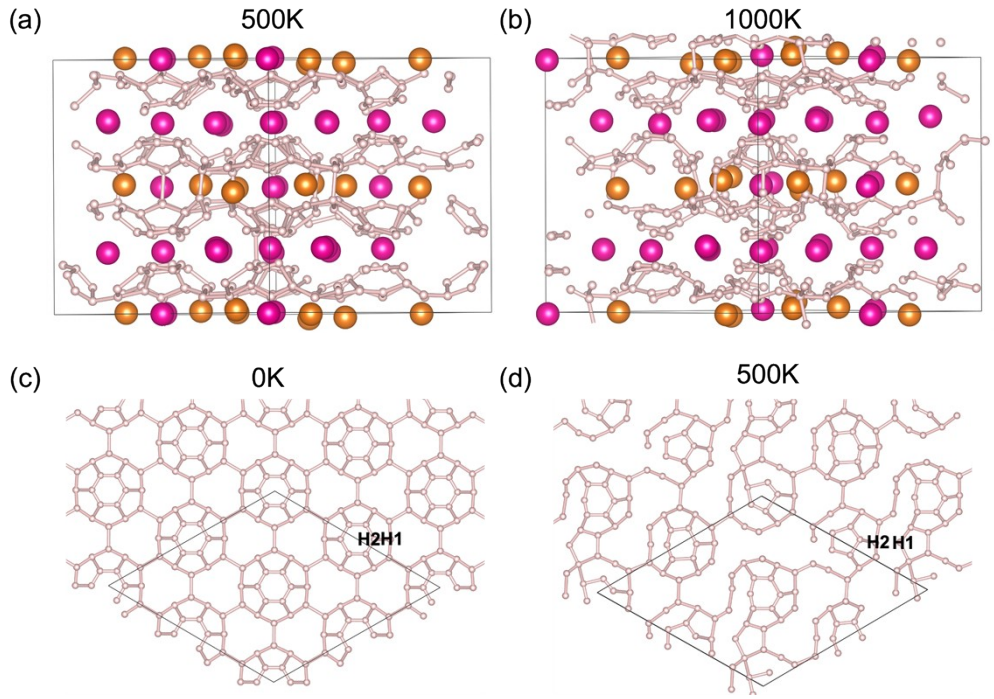

Fig. S7 Snapshots around 4000 steps of *P*6/*mmm* structure in the MD simulations under (a) 500 and (b) 1000 K. H layers in the *P*6/*mmm* structure at (c) 0 K and (d) 500 K. Two specific H were labeled as H1 and H2 to indicate the diffusion behavior of protons.

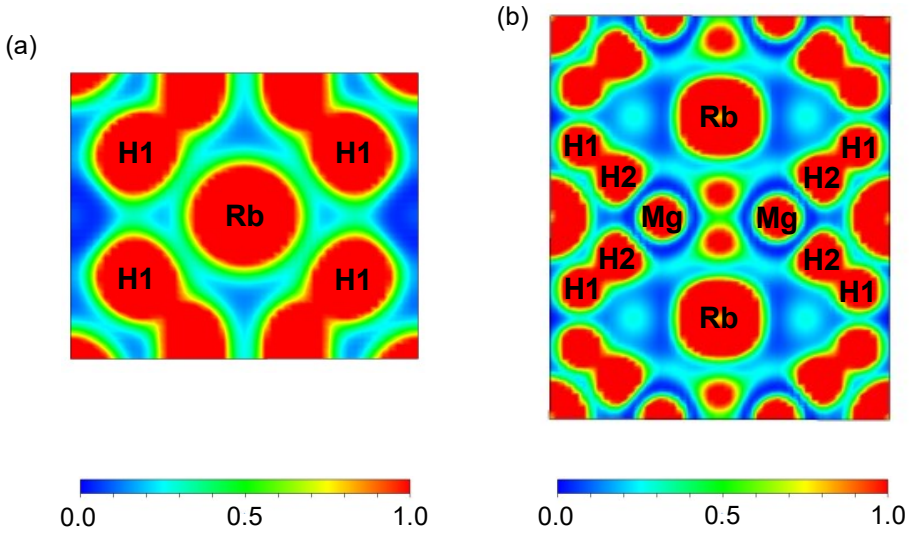

Fig. S8 2D sections of ELF on the (a) $(1\bar{1}0)$ and (b) (110) planes in Rb_2MgH_{18}-*P*6/*mmm* structure at 235 GPa.

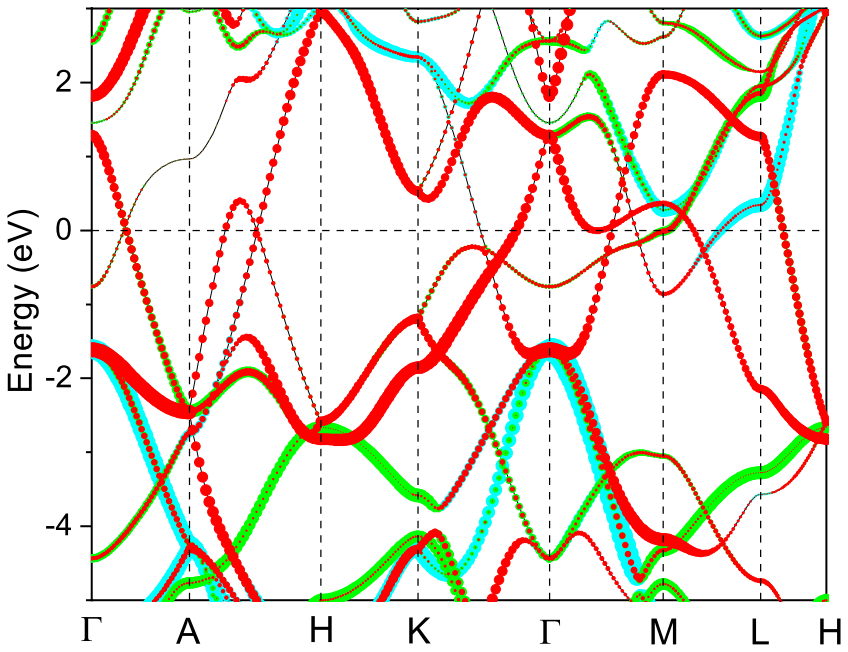

Fig. S9 Atom-projected band structure for H1, H2 and H3, which are indicted by cyan, green and red colors, respectively.

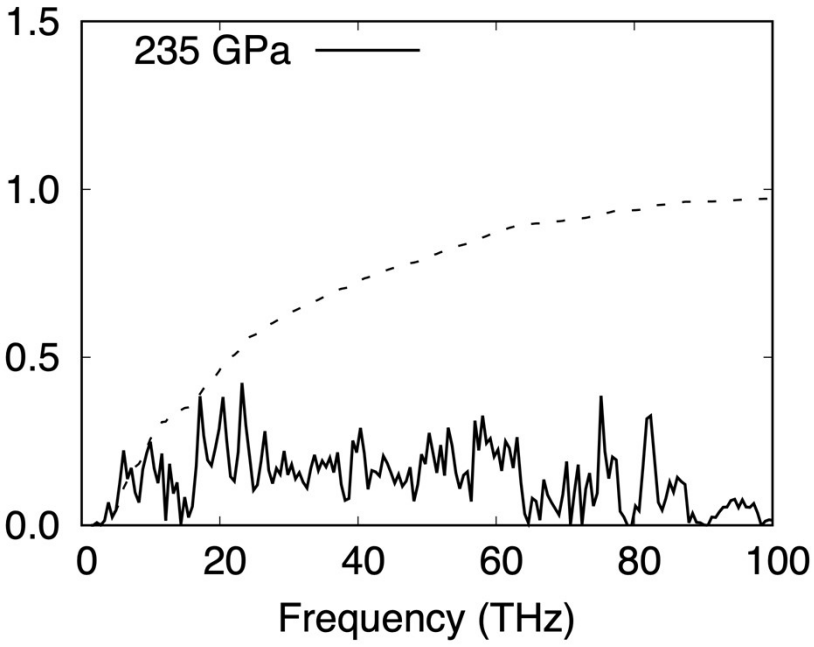

Fig. S10 Eliashberg spectral functions $\alpha^2F(\omega)$ (solid line) and integrated EPC constant (λ) (dashed line) of Rb_2MgH_{18}-$P1$ structure in a unit cell at 235 GPa.

Reference:

1 G. Kresse and D. Joubert, *Phys. Rev. B*, 1999, **59**, 1758–1775.

2 J. Sun, A. Ruzsinszky and J. P. Perdew, *Phys. Rev. Lett.*, 2015, **115**, 036402.