Supplementary Material for

**New Alkaline Earth Aluminum Hydride with One-Dimensional Zigzag Chains of $[\text{AlH}_6]$:
Synthesis and Crystal Structure of BaAlH_5**

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Structural refinement of BaAlD_5

The Rietveld refinement of BaAlD_5 was first carried out from the X-ray diffraction data of the deuteride sample using a program RIETAN-97. Figure S1 shows the observed and calculated X-ray diffraction patterns of the deuteride sample. The refined position coordinates of metal atoms and isotropic thermal displacement parameters for BaAlD_5 are listed in Table S1. Then the neutron powder diffraction data of the deuteride sample were analyzed by a Rietveld refinement program RIETAN-2001T. For the refinement, the initial Ba and Al atom coordinates were taken from those refined on the X-ray diffraction data and the deuterium positions were located by placing the atoms in the tetrahedral and octahedral holes formed by Ba and Al atoms. The occupation factors were also refined initially, but all the refined occupation factors were close to 1 (the differences were smaller than 3σ). Thus the occupation factors were fixed at unity for the final refinement. The finally refined position coordinates and isotropic thermal displacement parameters of BaAlD_5 are summarized in Table S2. The refined cell parameters and position coordinates of metal atoms are in good agreement with those refined from X-ray diffraction data within the range of 3σ (σ indicates the standard deviation).

Figure S1. The observed and calculated X-ray diffraction patterns of the deuteride sample. Reflection markers are for BaAlD_5 and Al (from above), respectively.

Table 1. Position coordinates of metal atoms and isotropic thermal parameters for BaAlD₅ refined from X-ray powder diffraction data^a

Atom	Site	<i>g</i>	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> (Å ²)
Ba	4a	1	0.6851(5)	0.150(1)	0.25 ^b	0.7(3)
Al	4a	1	0.051(2)	0.843(4)	0.234(1)	2.8(1)

^a Space group *Pna*2₁ (No. 33); cell parameters: *a* = 9.190(1) Å, *b* = 7.0399(5) Å, *c* = 5.1070(4) Å, *V* = 330.40(4) Å³, *Z* = 4; *R*_{wp} = 8.71%, *R*_p = 6.59%, *R*_i = 3.57%, *S* = 3.18. ^b This coordinate was constrained to fix the origin.

Table 2. Atomic coordinates and isotropic thermal parameters for BaAlD₅ refined from neutron powder diffraction data^a

Atom	Site	<i>g</i>	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> (Å ²)
Ba	4a	1	0.6873(7)	0.156(1)	0.25 ^b	0.5(1)
Al	4a	1	0.049(1)	0.847(2)	0.233(6)	4.5(3)
D1	4a	1	0.006(1)	0.939(1)	0.919(3)	2.7(2)
D2	4a	1	0.576(1)	0.846(1)	0.019(4)	2.6(2)
D3	4a	1	0.5720(9)	0.805(1)	0.497(4)	2.1(2)
D4	4a	1	0.3533(8)	0.696(1)	0.240(4)	3.2(2)
D5	4a	1	0.7112(8)	0.541(1)	0.209(2)	4.2(2)

^a Space group *Pna*2₁ (No. 33); cell parameters: *a* = 9.194(1) Å, *b* = 7.0403(9) Å, *c* = 5.1061(6) Å, *V* = 330.53(7) Å³, *Z* = 4; *R*_{wp} = 4.95%, *R*_p = 3.85%, *R*_i = 6.97%, *S* = 3.10. ^b This coordinate was constrained to fix the origin.