Supplementary Information for Deep Learning Generative Model for Conditional Crystal Structure Prediction of Sodium Amide

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Contents

Dataset construction and feature encoding

The custom dataset used for model training consists of 1000 candidate $NaNH_2$ structures generated using the AIRSS structure prediction method within the experimentally indexed lattice parameters. Each structure contains 64 atoms (16 $NaNH_2$ formula units) and was decomposed into a direct-space asymmetric unit (DAU). To determine the DAU substructure, we used `pymatgen`[1] to identify all symmetry-equivalent atomic groups in each structure. We then selected the group containing sodium atoms with the lowest site index to ensure a consistent and unique encoding for all structures. Each subunit in the DAU was encoded using six degrees of freedom: three Cartesian translations and three Euler angles defining position and orientation. These subunit descriptors were concatenated with the six lattice parameters (a, b, c, α, β, γ) to form an observed vector $\mathbf{z}$. To associate each structural configuration with a thermodynamic property, total energies E were computed using first-principles calculations. The resulting ($\mathbf{z}$, E) pairs formed the training data of both noise and energy models in the generative framework.

Neural network architecture and training

Two separate neural networks were used: a noise prediction model based on the Denoising Diffusion Probabilistic Model (DDPM),[2] and an energy regression model implemented as a multilayer perceptron (MLP). The noise model comprises three hidden layers of 64 units each, while the energy model uses three layers of 128 units, reflecting the increased complexity of the energy-structure relationship. Each layer uses ReLU activation functions to introduce non-linearity. All weights were initialized using the He uniform initialization strategy, which is well-suited for ReLU-based architectures and promotes stable convergence during training. All models were trained with a batch size of 32 using the Adam optimizer.[3] A `ReduceLROnPlateau` scheduler was applied to dynamically adjust the learning rate from an initial value of 1×10^{-4} to a minimum of 1×10^{-5}, with a decay factor of 0.6 and a patience of 20 epochs. Mean-squared error was used as the loss function for both models: the noise model

learns to recover the added Gaussian noise ε at each timestep, while the energy model minimizes the deviation from DFT-calculated total energies.

Diffusion sampling with energy guidance

Structure generation is initialized by sampling a noisy vector $\mathbf{z}_T \sim N(0, \mathbf{I})$ from a standard Gaussian distribution. This vector is iteratively denoised using the trained DDPM model, progressing through T discrete steps to recover a physically meaningful vector $\mathbf{z}_0$. At each step, the model estimates the residual noise, which is subtracted from the current latent vector to produce the next-step estimate. This iterative denoising process reconstructs the original structure parameterization from a highly perturbed latent representation. To improve the physical relevance of the generated structures, energy guidance is incorporated during sampling. At each denoising step, the energy model predicts the total energy corresponding to the current latent vector and computes its gradient with respect to $\mathbf{z}_t$. This gradient is used to introduce a deterministic correction, guiding the generation process toward energetically favorable regions.

Structure decoding and symmetry expansion

The denoised latent vector $\mathbf{z}_0$ is decoded to reconstruct the molecular configuration of the DAU. Each DAU consists of rigid $NaNH_2$ subunits, whose positions and orientations are determined by the translation and rotation parameters embedded in $\mathbf{z}_0$. The NH_2 group within each subunit is treated as a rigid fragment, based on its optimized DFT geometry with an N–H bond length of 1.03 ± 0.02 Å and an H–N–H bond angle of 104 ± 3°. To construct the full crystal structure, space group–specific Wyckoff symmetry operations are applied to the DAU. The decoding process accommodates different Wyckoff multiplicities by dynamically adjusting the number of subunits in the DAU, ensuring that the resulting atomic configuration conforms to the target crystallographic symmetry.

Post-processing and validation

To ensure structural validity, each generated structure is subjected to a two-stage post-expansion filtering process. First, a geometric filter checks all interatomic distances against predefined, element-specific thresholds to remove unphysical configurations. The minimum image convention is used to account for periodic boundary effects. Second, symmetry consistency is verified using the `spglib`[4] library. The symmetry matching is conducted with a distance tolerance `symprec` set to 1×10^{-2} Å, which enforces strict agreement between the generated structure and the assigned space group. Only structures that satisfy both criteria are retained for further analysis.

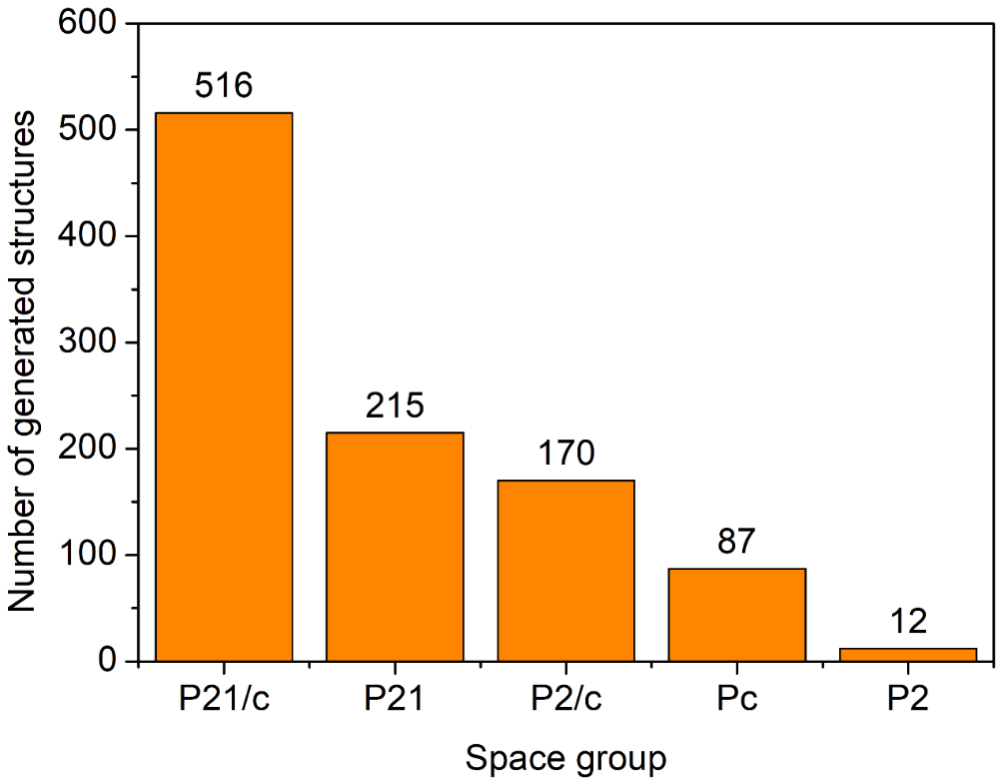
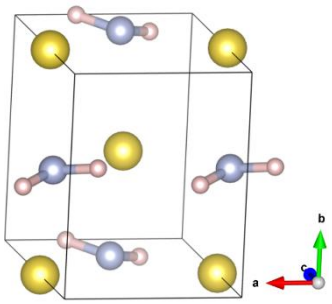


Fig. S1 Distribution of space groups in the generated $NaNH_2$ dataset.

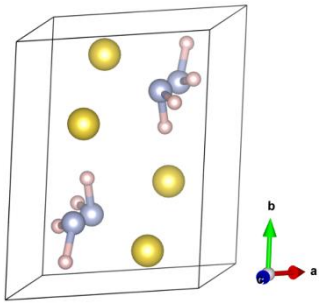
Table S1 Cell parameters (*a*, *b*, *c* in Å; α, β, γ in degrees) and Wyckoff positions for the unit cells of the top-ranked candidate structures of $NaNH_2$. These structures are generated using the conventional crystal structure prediction methods and optimized using DFT calculations at ambient pressure.

$P2_12_12$ ($Z = 2$)



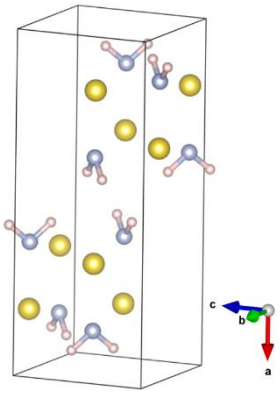
a, *b*, *c*	3.711	4.710	5.306
α, β, γ	90.000	90.000	90.000
H(4c)	0.2910	0.9615	0.8463
N(2b)	0.5000	1.0000	0.7208
Na(2a)	−0.0000	0.0000	0.4111

$P2_1/c$ ($Z = 4$)



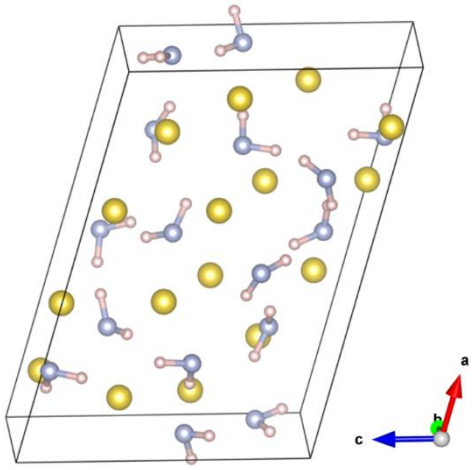
a, *b*, *c*	7.121	6.337	7.030
α, β, γ	90.000	145.882	90.000
H(4e)	0.1624	0.2712	0.4787
H(4e)	0.2581	0.3878	0.7556
N(4e)	0.2806	0.2429	0.7070
Na(4e)	0.3120	0.8606	0.7081

Pcca ($Z = 8$)



a, *b*, *c*	12.633	5.059	5.069
α, β, γ	90.000	90.000	90.000
H(8f)	1.4221	0.3611	1.1105
H(8f)	1.4215	0.1370	0.8893
N(8f)	1.3691	0.2496	1.0002
Na(8f)	1.3243	0.2502	1.5003

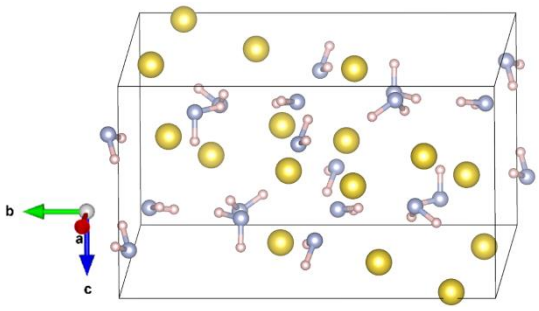
$P2_1$ ($Z = 16$)



a, *b*, *c*	12.806	6.306	9.096	Na(2a)	0.3763	0.2690	−0.0193
α, β, γ	90.000	106.029	120.000	Na(2a)	1.2062	0.9290	1.2242
H(2a)	0.2218	−0.0571	0.8647	Na(2a)	0.6238	0.7676	0.3668
H(2a)	0.1780	0.1199	0.9624	Na(2a)	0.3879	0.7327	0.4634
H(2a)	0.0348	0.2261	0.3493	Na(2a)	0.1249	0.3273	0.6860
H(2a)	−0.0452	0.4134	0.3726	Na(2a)	0.0973	0.7006	0.4213
H(2a)	1.0169	0.9439	0.0705	Na(2a)	0.6094	0.2856	0.8889
H(2a)	0.9871	0.7063	0.1082	Na(2a)	0.8353	0.1073	0.0411
H(2a)	0.6399	0.0893	0.1698				
H(2a)	0.7484	0.1335	0.3025				
H(2a)	0.8138	0.5886	0.5211				
H(2a)	0.7410	0.4366	0.3878				
H(2a)	0.4107	1.0895	0.3716				
H(2a)	0.4968	1.1091	0.2709				
H(2a)	0.4026	0.9374	0.8458				
H(2a)	0.5021	0.9491	0.7709				
H(2a)	0.7923	0.9853	0.7475				
H(2a)	0.6713	0.9288	0.7519				
N(2a)	0.2340	−0.0026	0.9758				
N(2a)	0.0282	0.3416	0.4273				
N(2a)	1.0014	0.8554	0.1585				
N(2a)	0.7153	0.0216	0.2188				
N(2a)	0.7500	0.4812	0.5005				
N(2a)	0.4756	1.0100	0.3492				
N(2a)	0.4774	1.0104	0.8620				
N(2a)	0.7255	1.0547	0.7722				

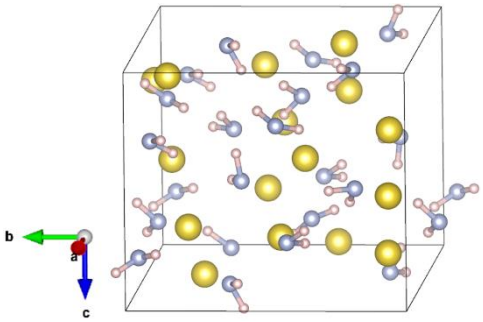
Table S2 Cell parameters (*a*, *b*, *c* in Å; α, β, γ in degrees) and Wyckoff positions for the unit cells of the top-ranked candidate structures of $NaNH_2$. These structures are generated using the proposed CSP model and optimized using DFT calculations at ambient pressure.

$P2_1/c$ ($Z = 16$)



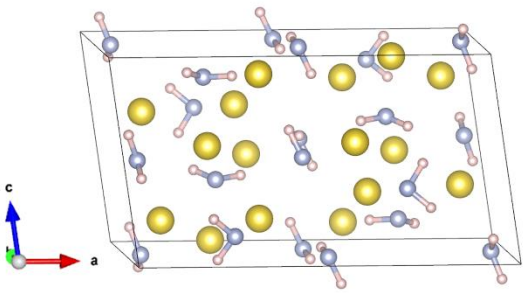
a, *b*, *c*	7.491	13.587	7.625
α, β, γ	90.000	72.452	90.000
H(4e)	0.3280	0.2476	0.2910
H(4e)	0.2342	0.3102	0.1600
H(4e)	0.3356	0.7582	0.2681
H(4e)	0.1626	0.8345	0.3004
H(4e)	0.1202	0.0027	0.3016
H(4e)	−0.0835	−0.0041	0.2728
H(4e)	0.5865	0.3874	0.7164
H(4e)	0.3667	0.4127	0.8038
N(4e)	0.2491	0.3104	0.2911
N(4e)	0.2082	0.7756	0.3628
N(4e)	0.0497	0.0236	0.2099
N(4e)	0.4962	0.4469	0.7549
Na(4e)	0.9527	0.3798	0.4857
Na(4e)	1.0224	0.6892	0.1630
Na(4e)	0.3443	0.0679	−0.0174
Na(4e)	−0.4582	0.3981	1.0756

$P2_1$ ($Z = 16$)



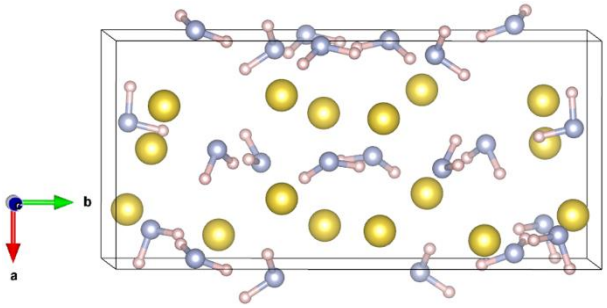
a, *b*, *c*	9.061	10.006	8.422
α, β, γ	90.000	79.733	90.000
H(2a)	0.0525	0.8840	0.7486
H(2a)	0.0325	0.0281	−0.1692
H(2a)	0.8650	0.6188	0.9232
H(2a)	0.7211	0.6169	0.0654
H(2a)	0.5983	1.0091	0.6581
H(2a)	0.4691	0.9918	0.8162
H(2a)	0.1320	0.3430	0.6260
H(2a)	−0.0341	0.3637	0.7247
H(2a)	0.3610	0.1638	0.4514
H(2a)	0.2265	0.1314	0.5983
H(2a)	0.6165	0.8376	0.2538
H(2a)	0.5903	0.9771	0.1598
H(2a)	0.8355	0.2941	0.5411
H(2a)	0.6711	0.2426	0.6322
H(2a)	0.3051	0.7987	0.8580
H(2a)	0.1584	0.7443	0.9734
N(2a)	−0.0299	0.9551	0.7853
N(2a)	0.7583	0.6598	0.9533
N(2a)	0.5577	0.9402	0.7477
N(2a)	0.0663	0.4153	0.6940
N(2a)	0.2566	0.1178	0.4747
N(2a)	0.5347	0.9065	0.2392
N(2a)	0.7647	0.2116	0.5508
N(2a)	0.2710	0.7208	0.9375
Na(2a)	0.2912	0.9839	0.2300
Na(2a)	0.2455	0.5964	1.1759
Na(2a)	0.7822	0.8026	0.7085
Na(2a)	0.5935	0.4097	0.4715
Na(2a)	0.7463	1.0890	0.3090
Na(2a)	0.8770	0.8582	0.0477
Na(2a)	−0.0072	0.0617	0.5173
Na(2a)	0.5210	0.7820	0.9783

*P*2/*c* (*Z* = 16)



a, *b*, *c*	13.228	7.554	7.341
α, β, γ	90.000	98.585	90.000
H(4g)	0.7784	0.6496	0.1180
H(4e)	0.6604	0.6117	0.1402
H(4e)	0.2139	0.0778	1.0248
H(4e)	0.2142	0.0047	0.2267
H(4e)	0.0392	0.2518	0.6001
H(4e)	1.0516	0.1786	0.4013
H(4e)	0.5090	0.3796	0.0433
H(4e)	0.5393	0.2581	−0.1163
N(4e)	0.7313	0.5490	0.1526
N(4e)	0.2655	0.0415	1.1395
N(4e)	0.9451	0.2971	0.0265
N(4e)	0.5152	0.2465	0.0117
Na(4e)	0.2426	0.7117	0.0404
Na(4e)	0.3863	−0.0204	0.4137
Na(4e)	0.3538	0.3238	0.1882
Na(4e)	0.1019	0.4302	0.1709

Pc (*Z* = 16)



a, *b*, *c*	6.682	14.273	7.511	Na(2a)	0.8417	0.4508	0.3081
α, β, γ	90.000	87.662	90.000	Na(2a)	0.2897	0.8980	0.4060
H(2a)	0.5527	0.2514	0.6482	Na(2a)	0.3451	0.4459	0.3948
H(2a)	0.6129	0.1891	0.8173	Na(2a)	0.2496	0.3480	0.7640
H(2a)	−0.1035	0.8621	0.3597	Na(2a)	0.4947	0.9129	1.0256
H(2a)	1.0066	0.7706	0.4343	Na(2a)	0.7794	0.0486	0.1589
H(2a)	0.0523	0.5101	0.5328	Na(2a)	0.6939	0.6570	0.4455
H(2a)	0.0962	0.6175	0.5859	Na(2a)	0.8850	0.7662	0.7643
H(2a)	1.1496	0.2825	0.4608				
H(2a)	0.9556	0.3256	0.5656				
H(2a)	0.9988	0.9357	0.9488				
H(2a)	0.8685	0.8587	1.0582				
H(2a)	0.4319	0.1199	0.2128				
H(2a)	0.2399	0.0531	0.2177				
H(2a)	0.6175	0.4002	0.6256				
H(2a)	0.5659	0.5114	0.6462				
H(2a)	0.5488	0.7081	0.8822				
H(2a)	0.4510	0.7087	0.6906				
N(2a)	0.4881	0.2157	0.7562				
N(2a)	0.9544	0.8359	0.4756				
N(2a)	0.0216	0.5782	0.4929				
N(2a)	1.0625	0.3426	0.4706				
N(2a)	0.8525	0.9120	0.9660				
N(2a)	0.3952	0.0489	0.2153				
N(2a)	0.5428	0.4474	0.7094				
N(2a)	0.5695	0.6791	0.7560				

Supplementary References

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