**Electronic Supplementary Information**

# Beryllium Dinitride Monolayer: A Multifunctional Direct Bandgap Anisotropic Semiconductor with Polymeric Nitrogen, Oxygen Reduction Catalysis, and Potassium Ion Storage Ability

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**Calculation of formation energy of α-2D-$BeN_2$**

The formation energy of α-2D-$BeN_2$ is calculated as:

$$E_{f-BeN_2} = \frac{E_{BeN_2} - E_{Be} - E_{N_2}}{3} \#(Eq\ S1)$$

where $E_{BeN_2}$, $E_{Be}$, and $E_{N_2}$ are the energy of α-2D-$BeN_2$, Be atom in its bulk metal and $N_2$ molecule, respectively.

**Calculation of the interlayer binding energy of the freestanding bilayers**

The interlayer binding energy of the freestanding bilayers is calculated as:

$$E_{bind} = \frac{E_{bi} - 2E_{mono}}{S} \#(Eq\ S2)$$

where $E_{bi}$, $E_{mono}$ and $S$ are the total energy of the bilayer, total energy of each monolayer, and the area of bilayer structure, respectively.

**Calculation of Young's stiffness and Poisson's ratio**

The in-plane Young's stiffness $Y(\theta)$ and Poisson's ratio $v(\theta)$ are calculated as functions of θ based on the equations as listed below [1]:

$$Y(\theta) = \frac{C_{11}C_{22} - C_{12}^2}{C_{11}sin^4\theta + C_{22}cos^4\theta + \left(\frac{C_{11}C_{22} - C_{12}}{C_{66}} - 2C_{12}\right)cos^2\theta sin^2\theta} \#(Eq\ S3)$$

$$v(\theta) = \frac{\left(C_{11} + C_{12} - \frac{C_{11}C_{22} - C_{12}^2}{C_{66}}\right)cos^2\theta sin^2\theta - C_{12}sin^4\theta - C_{12}cos^4\theta}{C_{11}sin^4\theta + C_{22}cos^4\theta + \left(\frac{C_{11}C_{22} - C_{12}}{C_{66}} - 2C_{12}\right)cos^2\theta sin^4\theta} \#(Eq\ S4)$$

**Calculation of carrier mobility**

According to the DP theory [2], the carrier mobility of a 2D structure is calculated as:

$$\mu_{2D} = \frac{e\hbar^3 C_{2D}}{k_B T m^* m_d (E_1)^2} \#(Eq\ S5)$$

where $e$, $\hbar$, and $k_B$ are the electron charge, reduced Planck constant, and Boltzmann constant, respectively. $C_{2D}$ are the elastic moduli, T is the temperature (300 K), $m^*$ is

the effective mass along the transport direction, which can be obtained by fitting the band structure at the CBM and VBM, and $m_d = \sqrt{m_x^* m_y^*}$ is the average effective mass. $E_l$ is the deformation potential constant, which is the key to the magnitude of mobility, defined by $E_l = \partial E_{edge}/\partial\varepsilon$, where $E_{edge}$ is the energy of the band edge and $\varepsilon = \Delta l/l_0$. The strain range is from -0.6% to +0.6% with an interval of 0.2%. This method has been widely applied in many monolayers [3-6].

**Calculation of optical absorption.**

According to the frequency-dependent permittivity of $\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$, the absorption coefficient was obtained based on the following equation [7, 8]:

$$\alpha(\omega) = \frac{\sqrt{2}\omega}{c}\left[\sqrt{\varepsilon_1^2(\omega) + \varepsilon_2^2(\omega)} - \varepsilon_1(\omega)\right]^{1/2} \#(Eq\ S6)$$

**Calculation of free energy changes for the oxygen reduction/evolution reaction (ORR/OER)**

The ORR and OER follow the associative mechanism pathways, as equation below:

$$O_2 \rightarrow OO* \rightarrow OOH* \rightarrow O* + H_2O \rightarrow OH* + H_2O \rightarrow 2H_2O \#(Eq\ S7)$$

$$2H_2O \rightarrow OH* + H_2O \rightarrow O* + H_2O \rightarrow OOH* \rightarrow OO* \rightarrow O_2 \quad \#(Eq\ 8)$$

where, the * is the α-2D-$BeN_2$.

To study the catalytic capabilities of α-2D-$BeN_2$, we employed a supercell combined with the computational hydrogen electrode (CHE) methodology [9]. The free energy (ΔG) of electrochemical reaction is expressed as:

$$\Delta G = \Delta E + \Delta E_{ZPE} - \Delta TS + \Delta G_{pH} + \Delta G_U \#(Eq\ S9)$$

where, ΔE represents the reaction energy; $E_{ZPE}$ and S respectively denote zero-point energy and entropy. The influences of pH and electrode potential (U) can be calculated by the equation as: ΔGpH = 0.0592 × pH and ΔG = −eU. In this study, the temperature (T) for the system is 298.15 K, and pH is 0 at an electrode potential (U) of 0 V. The limiting potential of ORR (UL,ORR) is defined as the minimum value among (−ΔG1, −ΔG2, −ΔG3, −ΔG4). The limiting potential of OER (UL,OER) is defined as the maximum value among (ΔG1, ΔG2, ΔG3, ΔG4). The $E_{ZPE}$/-TS of $H_2$ and $H_2O$(l) are 0.28/-0.404 eV and 0.68/-0.216 eV at 298.15 K and 1 atm obtained from the NIST database,10 and the Gibbs free energy of $H_2$ and $H_2O$(l) are calculated as -14.52 eV and -6.90 eV, respectively.

**Calculation details of K-ion storage performances**

The adsorption energy ($E_{ads}$) was calculated as following:

$$E_{ads} = E_{K@\alpha-2D-BeN_2} - E_{\alpha-2D-BeN_2} - E_K \#(Eq\ S10)$$

where $E_{K@\alpha-2D-BeN_2}$ and $E_{\alpha-2D-BeN_2}$ are total energy of adsorbed and bare α-2D-$BeN_2$, respectively. $E_K$ is the energy of a K atom in K metal bulk.

The formation energy ($E_f$) is calculated by the following equation:

$$E_f = \frac{E_{K_xBeN_2} - xE_K - E_{BeN_2}}{x+1} \#(Eq\ S11)$$

where $E_{K_xBeN_2}$ represents the energy of the system with x K ions.

The average stepwise adsorption energy ($E_{step}$) was calculated as following:

$$E_{step} = \frac{E_{K_nBeN_2} - E_{K_mBeN_2}}{n-m} - E_K \#(Eq\ S12)$$

where $E_{K_nBeN_2}$ and $E_{K_mBeN_2}$ are total energy of α-2D-$BeN_2$ with n and m K ions, respectively.

The maximum specific capacities ($C_M$) is estimated using the following formula:

$$C_M = \frac{cxF}{m_{\alpha-2D-BeN_2}} \#（Eq\ S13）$$

where c is the number of valence electrons (c = 1 for K), x is the number of K ions adsorbed per formula unit, F is the Faraday constant (26801 mAh·mol$^{-1}$), and $m_{\alpha-2D-BeN_2}$ is the molar weight of chemical formula $BeN_2$.

The open-circuit voltage (VOC) of these stable systems can then be calculated using the following formula:

$$V_{OC} = -\frac{E_{step}}{e} \#(Eq\ S14)$$

where the $E_{step}$ is the average stepwise adsorption energy between the stable adsorption conformers and e is the elementary charge.

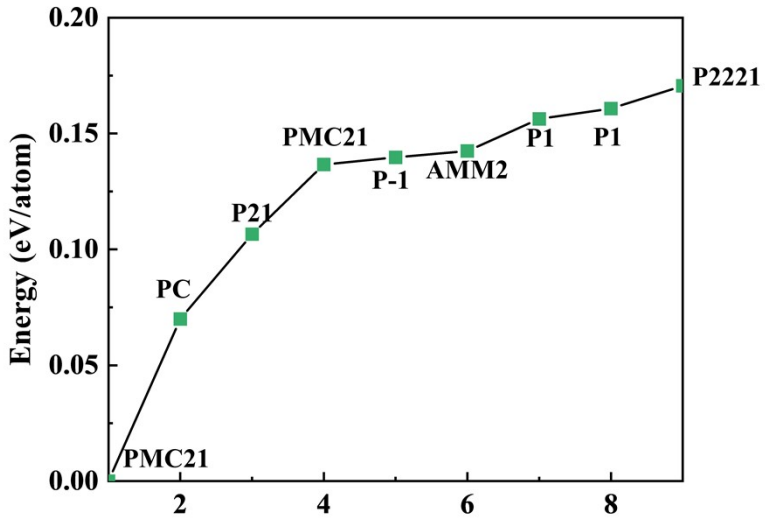


**Fig. S1.** The energy difference between α-2D-$BeN_2$ and 2D isomers of $BeN_2$, which are found by the CALYPSO code with atom number set as Be : N = 4 : 8.

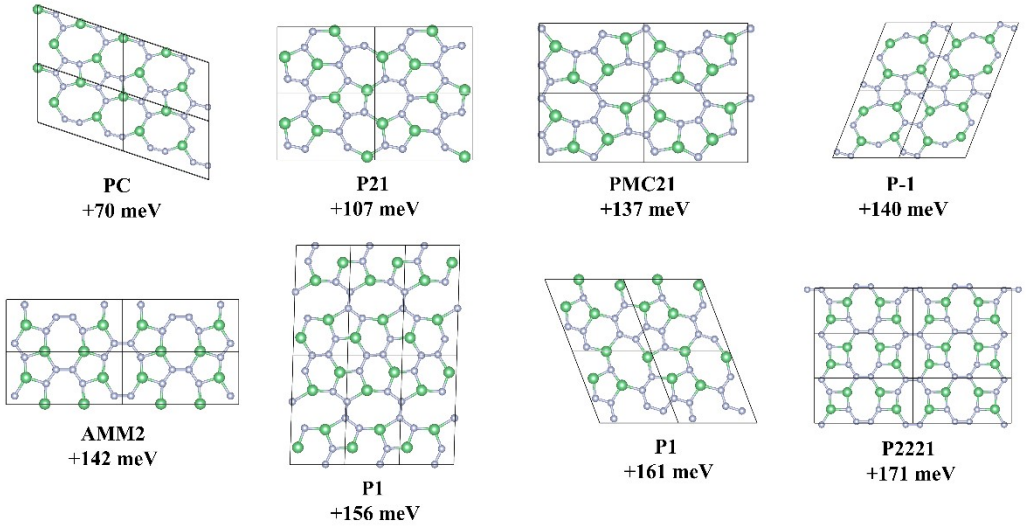


**Fig. S2.** The α-2D-$BeN_2$ and isomers of 2D $BeN_2$ found by the CALYPSO structure search code, where the number indicate the energy difference based on α-2D-$BeN_2$ as a reference.

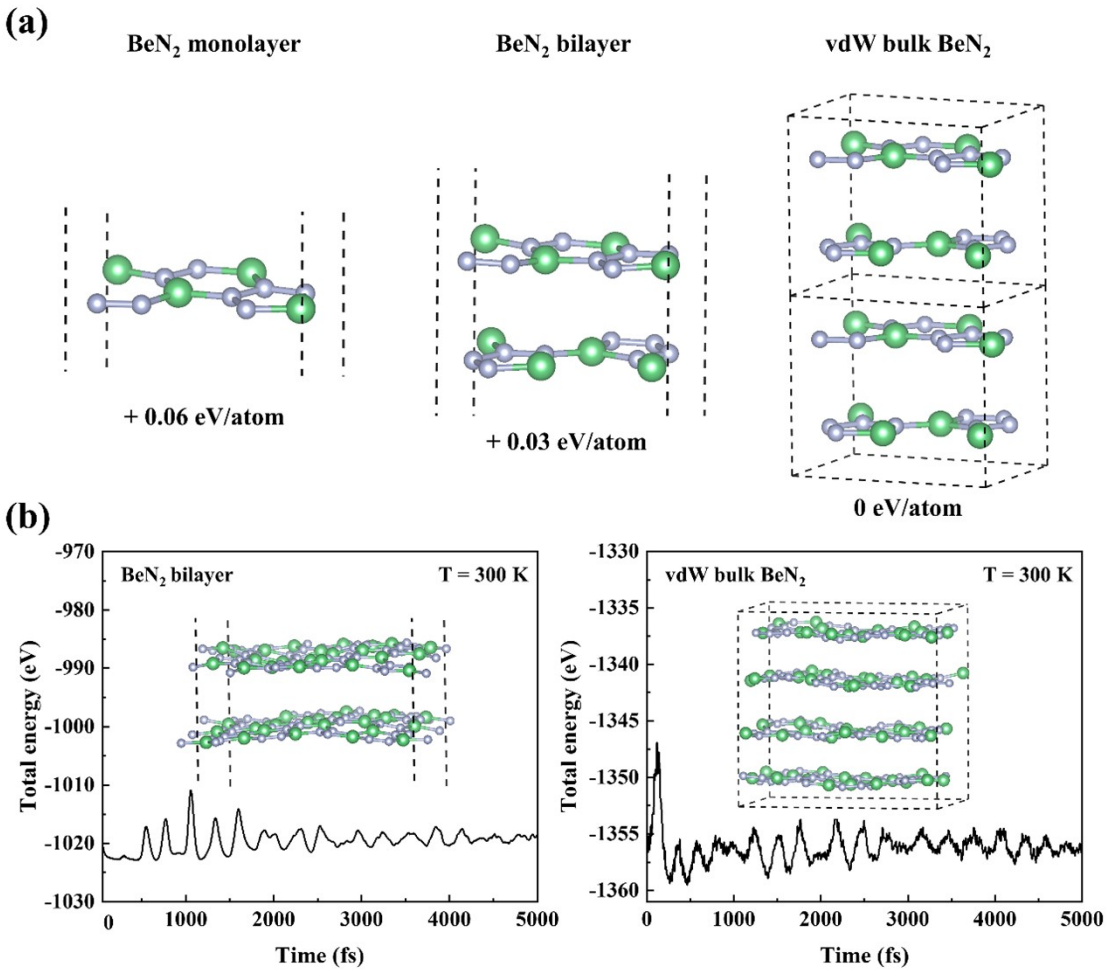


**Fig. S3.** (a) Structural configurations and relative energies (vs. vdW bulk $BeN_2$) of α-2D-$BeN_2$ monolayer, bilayer, and bulk phases. (b) the changes in total energy of α-2D-$BeN_2$ bilayer, and vdW bulk $BeN_2$, along with structural snapshots after the 5 ps AIMD simulations at 300 K. The supercell used in the AIMD simulation is 2 × 3 × 1, and 2 × 2 × 2, respectively.

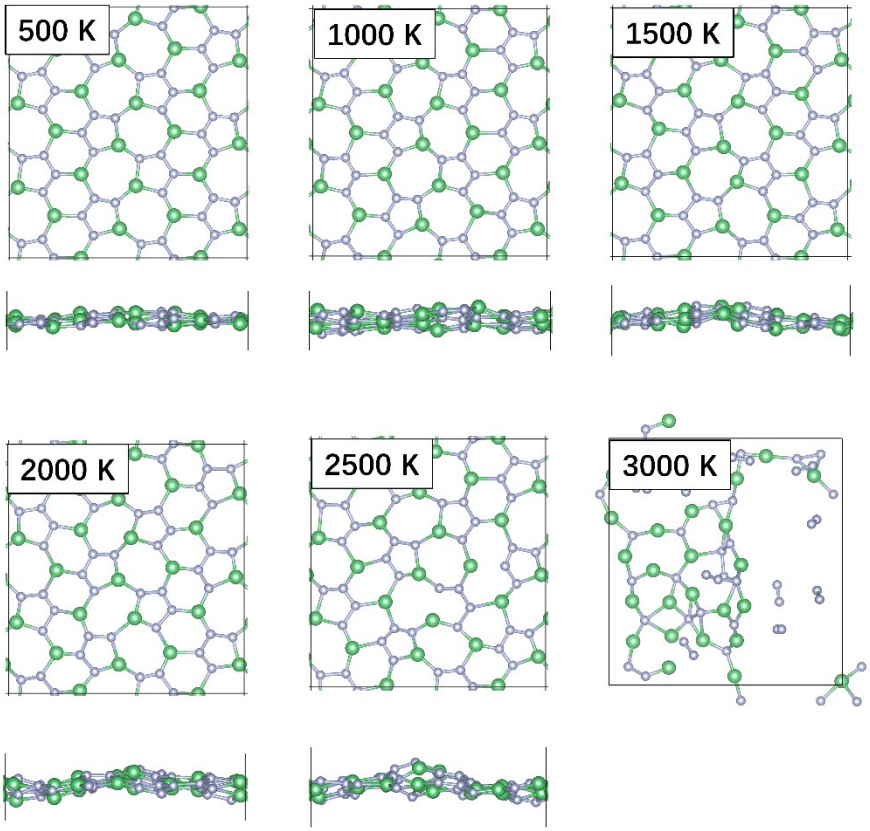


**Fig. S4.** The snapshots of α-2D-$BeN_2$ structures after AIMD simulations

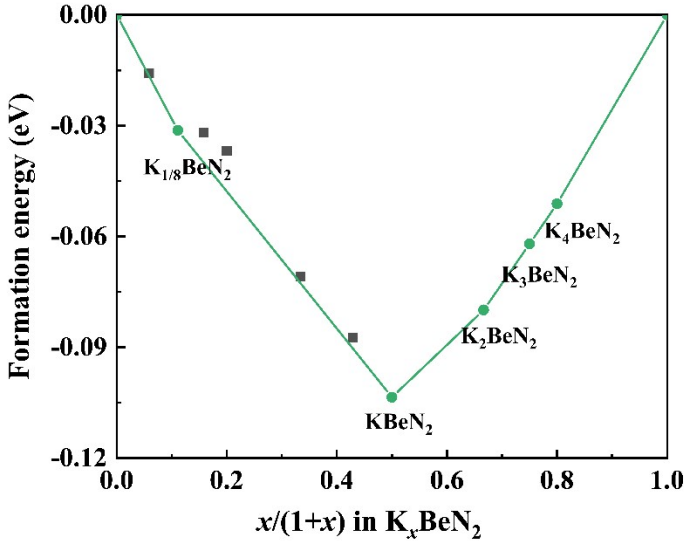


**Fig. S5.** Formation energy convex hull of $K_xBeN_2$ structures at different K ion concentrations.

**The carrier mobilities of α-2D-$BeN_2$**

The calculated details of all parameters mentioned above in the **"Calculation of carrier mobility"** part are listed in the Table S1. For electron carriers, the effective mass is 0.96/0.61 $m_e$ with high mobility of 0.55/6.6 × $10^4$ $cm^2V^{-1}s^{-1}$ along direction-*a*/-*b*, respectively. With comparison, the hole carrier has close effective mass of -0.60/-0.81 $m_e$ with relatively smaller mobility of 1.3/8.3 × $10^3$ $cm^2V^{-1}s^{-1}$, along direction-*a*/-*b*, respectively. The highest value is lower than that of graphene (3.4 × $10^5$ $cm^2V^{-1}s^{-1}$) [10] and *h*-$BeN_2$ (3.4 × $10^5$ $cm^2V^{-1}s^{-1}$) [4], but higher than that of BP monolayer (2.6 × $10^4$ $cm^2V^{-1}s^{-1}$) [5].

**Table. S1.** The calculated 2D elastic moduli ($C_{2D}$)，effective mass (m*), average effective mass ($m_d$), deformation potential constant ($E_l$) , and carrier mobility of 2D structures ($\mu_{2D}$) along the transport direction (axial ***a*** and ***b***, here). The "$m_e$" is the rest mass of a static electron.

| Carrier type | $C_{2D}$ (N/m) | $m^*$ ($m_e$) | $m_d$ ($m_e$) | $E_l$ (eV) | $\mu_{2D}$ ($cm^2V^{-1}s^{-1}$) |
|---|---|---|---|---|---|
| **Hole_a** | 153.8 | -0.600 | 0.698 | -2.414 | 1342 |
| **Electron_a** | 153.8 | 0.963 | 0.764 | -0.896 | 5546 |
| **Hole_b** | 145.4 | -0.813 | 0.698 | -0.813 | 8256 |
| **Electron_b** | 145.4 | 0.606 | 0.764 | -0.318 | 66143 |

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