Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

ReaxFF-MPNN Machine Learning Potential: A Combination of Reactive Force Field and Message Passing Neural Networks

Li-Yuan Xue [a)], Feng Guo[a)b)1], Yu-Shi Wen[c)2], Shi-Quan Feng[d)], Xiao-Na Huang [e)], Lei Guo[f)], Heng-Shuai Li[a)], Shou-Xin Cui[b)], Gui-Qing Zhang [b)], and Qing-Lin Wang[a)b)]

[a)]*Shandong Provincial Key Laboratory of Optical Communication Science and Technology, Liaocheng 252000, China*

[b)]*School of Physical Science and Information Technology, Liaocheng University, Liaocheng 252000, China*

[c)]*Institute of Chemical Materials, China Academy of Engineering Physics (CAEP), Mianyang, Sichuan 621900, China*

[d)]*School of Physics and Electronic Engineering, Zhengzhou University of Light Industry, Zhengzhou 450002, China*

[e)] *Department of Mechanical Engineering, City University of Hong Kong, 83 Tat Chee Avenue, Hong Kong*

[f)]*School of Business, Shandong Normal University, Jinan, 250014, China*

-Supporting Information-

1gfeng.alan@gmail.com

2wenys@caep.cn

1. Energy terms used in *ReaxFF-MPNN*

We use bond energy, three-body bond angle energy, four-body torsion energy, and long-range energy such as H-bond energy, van der Waals energy, and Coulomb energy. The over/under coordinate and lone pair energy terms are not used in the ReaxFF-MPNN as shown in (s1).

$$E_{total} = E_{bond} + E_{val} + E_{pen} + E_{coa} + E_{tors} + E_{conj} + E_{H-bond} + E_{vdWaals} + E_{C} \quad \text{(s1)}$$

2. Bond energy in *ReaxFF-MPNN*

We have considered two forms of bond energy formula and the two kinds of bond energy in ReaxFF-MPNN are expressed by formula (s2) and (s3).

$$E^{bond}_{ij} = -D^{\sigma}_{e} \cdot f^{E_{bond}}_{NN}\left(BO^{\sigma}_{ij}, BO^{\pi}_{ij}, BO^{\pi\pi}_{ij}\right) \cdot BO_{ij} \quad \text{(s2)}$$

$$E^{bond}_{ij} = -D^{\sigma}_{e} \cdot f^{E_{bond}}_{NN}\left(\Delta_i - BO_{ij}, \Delta_j - BO_{ij}, BO_{ij}\right) \cdot BO_{ij} \quad \text{(s3)}$$

Where D^{σ}_{e} is adjusting parameters, and $f^{E_{bond}}_{NN}$ is a neural network. Typically, a neural network function can be expressed by (s4).

$$f_{NN}(X) = \sigma(W \cdot \cdots \sigma(W \cdot \sigma(W \cdot X + b) + b) + \cdots + b) \quad \text{(s4)}$$

σ represents the activation function, and in this work, we use the sigmoid activation function as in formula (s5).

$$\sigma(X) = \frac{1}{1 + e^{-X}} \quad \text{(s5)}$$

The X represents the input vector, W and b represent the *weight* and *bias* of the neural network.

3. Bond order in *ReaxFF-MPNN*

The initial bond order is $BO^{t=0}\left(BO^{t=0}_{\sigma} + BO^{t=0}_{\pi} + BO^{t=0}_{\pi\pi}\right)$ which is computed from ReaxFF uncorrected bond order $BO'\left(BO'_{\sigma} + BO'_{\pi} + BO'_{\pi\pi}\right)$ with neural network functions.

$$BO^{t=0}_{\sigma} = f^{\sigma}_{NN}\left(BO'_{\sigma}\right) \quad \text{(s6)}$$

$$BO^{t=0}_{\pi} = f^{\pi}_{NN}\left(BO'_{\pi}\right) \quad \text{(s7)}$$

$$BO^{t=0}_{\pi\pi} = f^{\pi\pi}_{NN}(BO'_{\pi\pi}) \tag{s8}$$

We use $BO^{t=1}(BO^{t=1}_{\sigma} + BO^{t=1}_{\pi} + BO^{t=1}_{\pi\pi})$ as the final bond state, the $BO^{t=1}$ is computed by formula (s9).

$$BO^{t=1}_{ij} = f^{message}_{NN}(\Delta^{t=0}_{i} - BO^{t=0}_{ij}, \Delta^{t=0}_{i} - BO^{t=0}_{ij}, BO^{t=0}_{ij}) \odot BO^{t=0} \tag{s9}$$

where $\odot$ denote the element multiplication, and Δ is calculated by the formula (s10).

$$\Delta_i = \sum_{j \in N_i} BO_{ij} \tag{s10}$$

4. The bond energy curve of the NN bond of structure $NO_2N(CH_3)_2$ and $NO_2NCH_3CH_2NH_2$ from DFT calculations

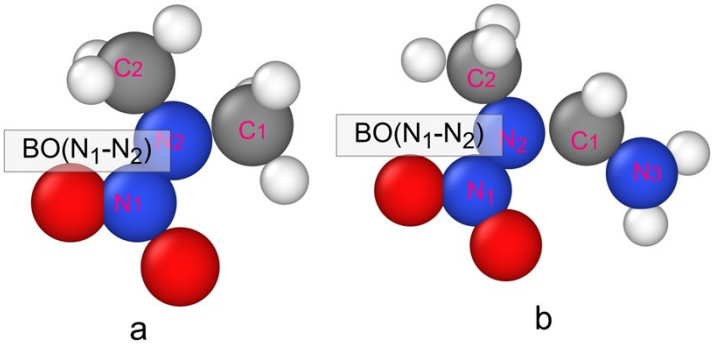

Figure s1. The structure of (a) $NO_2N(CH_3)_2$ and (b) $NO_2NCH_3CH_2NH_2$.

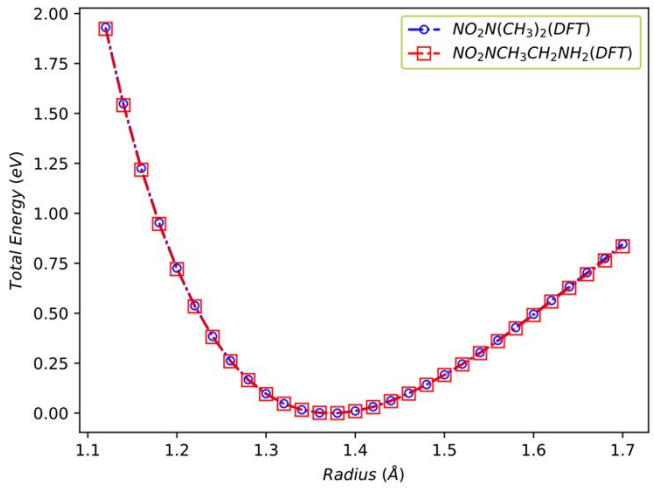

Figure s2. The bond energy versus distance between N_1-N_2 of the structure of $NO_2N(CH_3)_2$ and $NO_2NCH_3CH_2NH_2$ calculated by SIESTA with PBE functional and double-ζ polarize basis set as shown in Figure s1(a) and Figure s2(b), respectively.

5. A neural network taper function for vdW interaction

ReaxFF uses a distance-corrected Morse-potential and a shielded interaction to provide nuclear repulsion. A 7th order polynomial taper correction function developed by de Vos Burchart is employed to avoid energy discontinuities. We have tested a new neural network taper function instead of the polynomial taper function with parameter adjustable (the new VDW function is given in formula s11). The comparison of neural network taper function and polynomial taper function is provided in Figure s3.

$$E_{vdWaals} = f_{NN}^{Taper} \cdot D_{ij} \cdot \left\{ exp\left[\alpha_{ij} \cdot \left(1 - f_{13}\frac{(r_{ij})}{r_{vdW}}\right)\right] - 2 \cdot exp\left[\frac{1}{2} \cdot \alpha_{ij} \cdot \left(1 - f_{13}\frac{(r_{ij})}{r_{vdW}}\right)\right]\right\} \quad \text{(s11)}$$

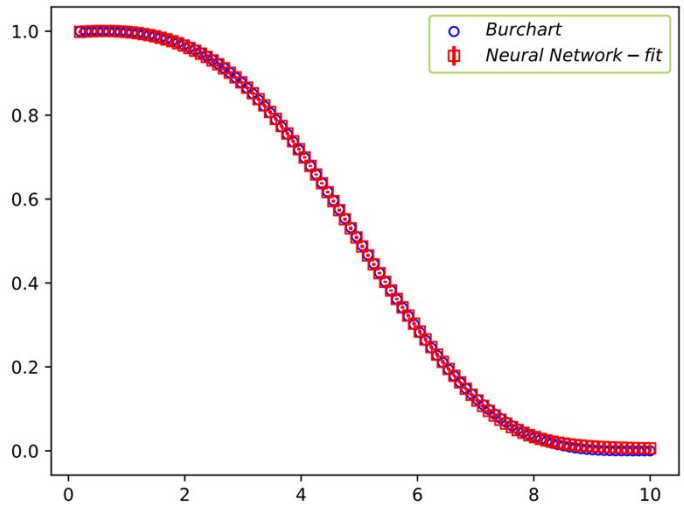

Figure s3. The plot of the ReaxFF taper function developed by Burchart et. al. (blue circle) and the neural network taper function (red square error bar).