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

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-Supporting Information-

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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).

\[ E_{total} = E_{bond} + E_{val} + E_{coa} + E_{tors} + E_{conj} + E_{H-bond} + E_{vdW} + E_C \]  
(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).

\[ E_{bond}^{ij} = -D_e \cdot f_{NN}^E(BO_\sigma^{ij}BO_\pi^{ij}BO_{\pi\pi}^{ij}) \cdot BO_{ij} \]  
(s2)

\[ E_{bond}^{ij} = -D_e \cdot f_{NN}^E(\Delta_i - BO_\sigma^{ij}\Delta_j - BO_\pi^{ij}BO_{ij}) \cdot BO_{ij} \]  
(s3)

Where \( D_e \) is adjusting parameters, and \( f_{NN}^E \) is a neural network. Typically, a neural network function can be expressed by (s4).

\[ f_{NN}(X) = \sigma(W \cdot \ldots \sigma(W \cdot \sigma(W \cdot X + b) + b) + \ldots + b) \]  
(s4)

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

\[ \sigma(X) = \frac{1}{1 + e^{-X}} \]  
(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 \( BO^t = (BO_\sigma^t + BO_\pi^t + BO_{\pi\pi}^t) \) which is computed from ReaxFF uncorrected bond order \( BO'(BO_\sigma' + BO_\pi' + BO_{\pi\pi}') \) with neural network functions.

\[ BO_\sigma^t = f_{NN}^\sigma(BO_\sigma') \]  
(s6)

\[ BO_\pi^t = f_{NN}^\pi(BO_\pi') \]  
(s7)
\[ BO_{\pi}^t = 0 = f_{\pi \pi}^{NN}(BO'_{\pi \pi}) \]  
\[(s8)\]

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).

\[ BO_{ij}^{t=1} = f_{\text{message}}^{NN}(\Delta_{i=0}^{t=0} - BO_{ij}^{t=0} \Delta_{i=0}^{t=0} - BO_{ij}^{t=0} BO_{ij}^{t=0}) \odot BO_{ij}^{t=0} \]  
\[(s9)\]

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

\[ \Delta_i = \sum_{j \in N_i} BO_{ij} \]  
\[(s10)\]

4. The bond energy curve of the NN bond of structure \( \text{NO}_2\text{N(CH}_3)_2 \) and \( \text{NO}_2\text{NCH}_2\text{CH}_2\text{NH}_2 \) from DFT calculations

Figure s1. The structure of (a) \( \text{NO}_2\text{N(CH}_3)_2 \) and (b) \( \text{NO}_2\text{NCH}_2\text{CH}_2\text{NH}_2 \).
Figure s2. The bond energy versus distance between N₁-N₂ of the structure of NO₂N(CH₃)₂ and NO₂NCH₃CH₂NH₂ 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.

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

(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).