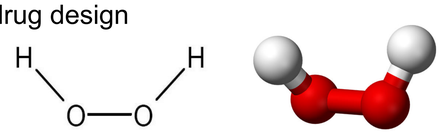
ComENet: Towards Complete and Efficient Message Passing for 3D Molecular Graphs

Limei Wang*[1], Yi Liu*[2], Yuchao Lin[1], Haoran Liu[1], Shuiwang Ji[1]
[1]Texas A&M University, [2]Florida State University

Introduction

Problem: representation learning for 3D molecular graphs

- Use ML to reduce the computation time required for predicting molecular properties
- For applications such as batteries, catalysis and drug design



- Molecules can be modeled as 3D graphs where each node has a spatial position in 3D.
- 3D positions contain essential physical information and is crucial for molecular modeling.

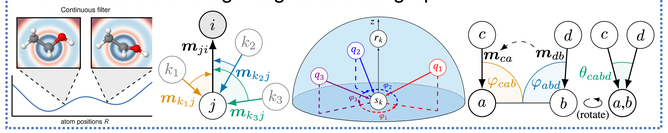
Challenges: how to incorporate 3D information completely and efficiently?

- Existing methods either use partial 3D information, or suffer from excessive computational cost.

Existing methods

	3D information	Complexity
SchNet [1]	Distances between atoms d	$O(nk)$
DimeNet [2]	d + Angles between edges θ	$O(nk^2)$
SphereNet [3]	d, θ + Angles between 4 nodes (2-hop) ϕ	$O(nk^2)$
GemNet [4]	d, θ + Angles between 4 nodes (3-hop) τ	$O(nk^3)$

n denotes the number of nodes
k denotes the average degree in a 3D graph



Proposed ComENet: complete and efficient graph neural network

- Complete:
  - Local completeness
  - Global completeness
  - With rigorous proof
- Efficient:
  - Reduce the complexity from $O(nk^2)$ or $O(nk^3)$ to $O(nk)$
  - By operates within 1-hop neighborhood
- Invariant to rotation and translation of input 3D graphs

Methodology

Notations:

A 3D graph $G = (V, A, P)$
Node feature matrix $V = [\mathbf{v}_1, \mathbf{v}_2, \dots, \mathbf{v}_n]^T \in \mathbb{R}^{n \times d_v}$
Adjacency matrix A
Position matrix $P = [\mathbf{p}_1, \mathbf{p}_2, \dots, \mathbf{p}_n]^T \in \mathbb{R}^{n \times 3}$

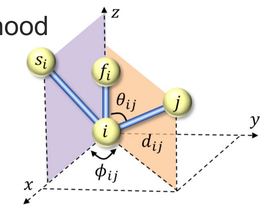
Definition (Completeness).

For two 3D graphs $G_1 = (V, A, P_1)$ and $G_2 = (V, A, P_2)$, a geometric transformation $\mathcal{T}$ is considered as complete when

$$\mathcal{T}(G_1) = \mathcal{T}(G_2) \Leftrightarrow \exists \mathcal{R} \in SE(3), P_1 = \mathcal{R}(P_2)$$

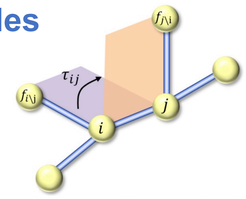
Local completeness

Local structure: a node and its 1-hop neighborhood
Define the local coordinate system for node i
f_i: i's nearest neighbor
s_i: i's second nearest neighbor
Define x, y, z-axis
Use spherical coordinates $(d_{ij}, \theta_{ij}, \phi_{ij})$ determine the relative position of each neighboring node j

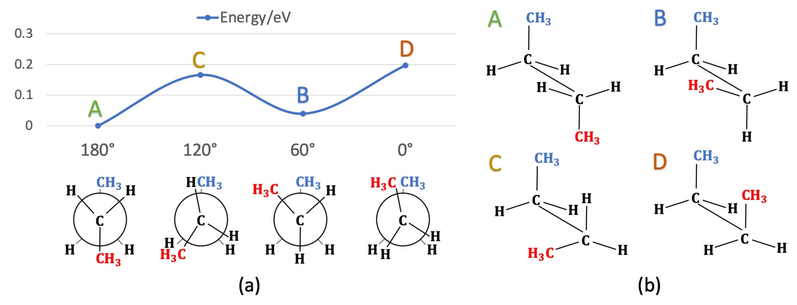


Global completeness via rotation angles

The only remaining degree of freedom is the rotation angles between local structures
Use edge rotation angles τ_{ij} to achieve global completeness



Rotation angles for conformer identification



(a) Illustration that the relative conformation energy of butane is a function of the rotation angle of the C-C bond. (b) A 3D view of the four conformers in (a).

Message passing scheme

$$\mathbf{v}_i^{l+1} = g\left(\mathbf{v}_i^l, \sum_{j \in \mathcal{N}_i} f\left(\mathbf{v}_j^l, d_{ij}, \theta_{ij}, \phi_{ij}, \tau_{ij}\right)\right)$$

g and f can be implemented by neural networks or mathematical operations.

Experiments

Datasets

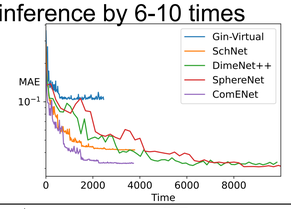
Two large-scale datasets [5,6] and one commonly used dataset [7]

Dataset	OC20 [5]	Molecule3D [6]	QM9 [7]
# Graphs	660,010	3,899,647	130,831
Split Type	Pre-defined	Random/Scaffold	Random
Split Ratio	70:15:15	6:2:2	84:8:8
# Nodes/Graph	77.75	29.11	18.02

Performance and efficiency

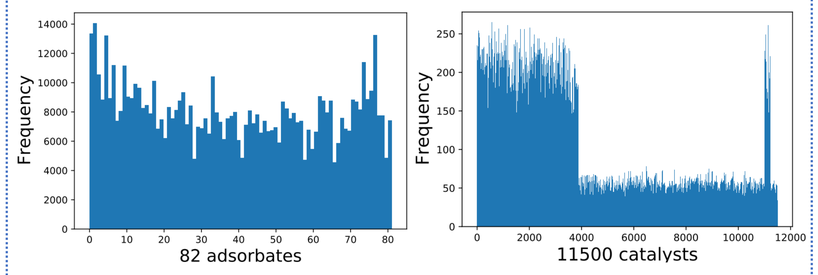
Achieve similar or better performance compared with existing methods
Dramatically accelerates the training and inference by 6-10 times

Model	Time		MAE	
	Train	Inference	Random	Scaffold
GIN-Virtual	15min	2min	0.1036	0.2371
SchNet	14min	3min	0.0428	0.1511
DimeNet++	133min	16min	0.0306	0.1214
SphereNet	182min	28min	0.0301	0.1182
ComENet	22min	3min	0.0326	0.1273



Model	Time		Energy MAE [eV] ↓					EwT ↑				
	Train	Infer.	ID	OOD Ads	OOD Cat	OOD Both	Average	ID	OOD Ads	OOD Cat	OOD Both	Average
CGCNN	18min	1min	0.6203	0.7426	0.6001	0.6708	0.6585	3.36%	2.11%	3.53%	2.29%	2.82%
SchNet	10min	1min	0.6465	0.7074	0.6475	0.6626	0.6660	2.96%	2.22%	3.03%	2.38%	2.65%
DimeNet++	230min	4min	0.5636	0.7127	0.5612	0.6492	0.6217	4.25%	2.48%	4.40%	2.56%	3.42%
GemNet-T	200min	4min	0.5561	0.7342	0.5659	0.6964	0.6382	4.51%	2.24%	4.37%	2.38%	3.38%
SphereNet	290min	5min	0.5632	0.6682	0.5590	0.6190	0.6023	**4.56%**	2.70%	**4.59%**	2.70%	**3.64%**
ComENet	20min	1min	**0.5558**	**0.6602**	**0.5491**	**0.5901**	**0.5888**	4.17%	**2.71%**	4.53%	**2.83%**	3.56%

Ablation Study for Identifying Conformers



Distributions of adsorbates and catalysts in OC20. For y-axis, frequency counts the number of conformers for each individual adsorbate and catalyst.

Model	Energy MAE [eV] ↓					EwT ↑				
	ID	OOD Ads	OOD Cat	OOD Both	Average	ID	OOD Ads	OOD Cat	OOD Both	Average
ComENet	**0.5558**	**0.6602**	**0.5491**	**0.5901**	**0.5888**	**4.17%**	**2.71%**	**4.53%**	**2.83%**	**3.56%**
ComENet w/o τ	0.5585	0.6851	0.5574	0.6186	0.6049	4.13%	2.65%	4.13%	2.75%	3.42%

Removing rotation angles τ can harm the performance of ComENet, demonstrating the effectiveness of our global complete representations for identifying conformers.

Our code is available at: https://github.com/divelab/DIG (Deep learning on 3D graphs)

Reference: *[1] Schütt, K., et al. "SchNet: A continuousfilter convolutional neural network for modeling quantum interactions." NeurIPS 2017. [2] Gasteiger, J., et al. "Directional message passing for molecular graphs." ICLR 2020. [3] Liu, Y., et al. "Spherical message passing for 3D molecular graphs." ICLR 2022. [4] Gasteiger, J., et al. "GemNet: Universal directional graph neural networks for molecules." NeurIPS 2021. [5] Chanussot, L., et al. "Open catalyst 2020 (OC20) dataset and community challenges." ACS Catalysis. [6] Xu, Z., et al. "Molecule3D: A benchmark for predicting 3D geometries from molecular graphs." [7] Ramakrishnan, R., et al. "Quantum chemistry structures and properties of 134 kilo molecules." Scientific data.*

Paper

Code