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Interatomic Potential Prediction Based on Charge Equilibration and Equivariant Transformer

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Abstract

Conventional local machine-learning interatomic potentials describe atomic environments with a finite cutoff radius and therefore have difficulty capturing long-range electrostatic coupling in polar, charged, or charge-transfer systems. This paper proposes CE-ETNet (Charge-Equilibration-Enhanced Equivariant Transformer Network), an equivariant Transformer interatomic potential constrained by charge equilibration. The model learns local chemical environments formed by atom types, geometric edges, and radial basis features through an equivariant representation encoder, and predicts atomic electronegativities. Under the constraint of total charge conservation, a differentiable charge equilibration procedure is used to solve partial charges and electrostatic energy, thereby incorporating long-range Coulomb interactions into energy and force prediction. On three benchmark datasets, QM9, revised MD17, and tmQM_wB97MV, CE-ETNet reduces the mean absolute error (MAE) of energy and force prediction by about 8% on average compared with existing models, reaching chemical accuracy. The experimental results show that explicit long-range electrostatic modeling improves the predictive performance of equivariant graph neural network interatomic potentials and provides a useful design direction for machine-learning interatomic potential models.

Keywords

interatomic potential, equivariant Transformer, graph neural network, machine learning

1. Introduction

Traditional molecular dynamics simulations usually rely on classical force fields or quantum-mechanical calculations. The former are computationally efficient, but their parameterized functional forms are difficult to generalize to complex chemical environments; the latter can provide highly accurate potential energies and forces, but at a much higher computational cost. In recent years, machine-learning interatomic potentials (MLIPs) have offered a promising route to balancing efficiency and accuracy by learning the mapping between atomic configurations and potential energy surfaces, and by obtaining atomic forces from the gradient of the potential energy with respect to atomic coordinates[1].

Most existing MLIPs take atom types, relative coordinates, interatomic distances, and local geometric relations as inputs, and extract atomic-environment features using graph neural networks (GNNs) or equivariant graph neural networks (EGNNs) [2-4]. Equivariant features allow the predicted energy to remain invariant under global rotation and translation, while the predicted forces transform equivariantly with

coordinates[5-6]. This makes such models well suited for learning potential energy surfaces and atomic forces. However, to control computational cost, most models construct local neighborhoods with a finite cutoff radius, so electrostatic coupling between distant atoms can only be fitted indirectly through local features.

For polar, ionized, or charge-transfer molecular systems, charge redistribution and Coulomb interactions have a substantial influence on the total energy and atomic forces. Without an explicit electrostatic description, local potential models may suffer from generalization errors when conformations or charge states change[2,7-9]. Therefore, introducing an electrostatic energy term that satisfies charge conservation on top of equivariant local features is a key problem for improving interatomic potential prediction.

To address this issue, this paper proposes CE-ETNet (Charge-Equilibration-Enhanced Equivariant Transformer Network), an equivariant Transformer interatomic potential with charge-equilibration constraints. The model uses an equivariant encoder to extract local geometric representations and predict environment-dependent atomic electronegativities. It then solves equilibrium charges and electrostatic energy under the total-charge constraint according to the charge equilibration principle. Finally, the electrostatic energy is combined with the short-range atomic energy, and atomic forces are obtained from the gradient of the total energy. Through these steps, local equivariant atomic features, charge conservation, and electrostatic-energy modeling are integrated into a unified differentiable computation, enabling accurate prediction of interatomic energies and forces in molecular systems.

2. Related Work

Existing machine-learning potentials usually represent atomic systems as graphs and learn atomic neighborhoods through message passing. SchNet generates continuous filters from interatomic distances and effectively describes distance-dependent local interactions, while DimeNet further introduces bond-angle information to make message passing more directionally expressive[3-4]. These methods mainly rely on finite cutoff neighborhoods, so their effective receptive fields are limited by the cutoff radius and the number of network layers.

Equivariant networks further incorporate geometric symmetry directly into model design. NequIP constructs E(3)-equivariant message passing with irreducible representations, spherical harmonics, and tensor products, achieving high accuracy in small-sample potential learning. PaiNN builds directional information through interactions between scalar and vector features. Allegro reduces the cost of high-order tensor operations by using local equivariant representations and achieves a balance between accuracy and efficiency[5-6,10]. Nevertheless, these models still mainly depend on local neighborhoods, and electrostatic interactions are usually learned implicitly by the network.

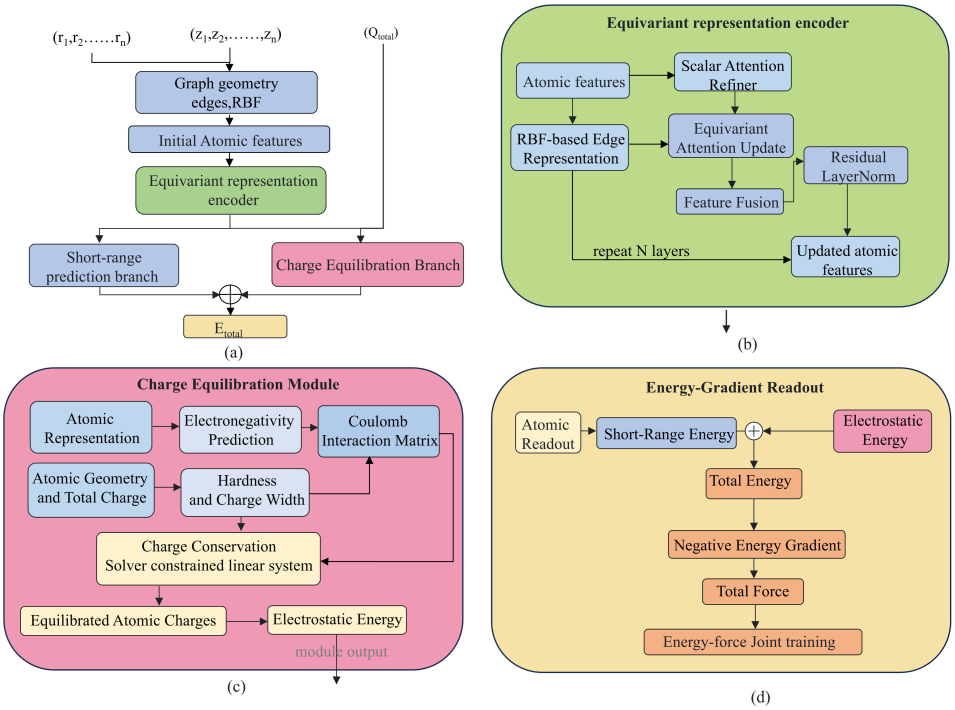
For long-range interactions, recent studies have begun to explicitly introduce charges or global information. PhysNet predicts partial charges from atomic representations and corrects atomic energies with screened Coulomb terms. SpookyNet combines global attention and electronic-state information to enhance nonlocal interaction modeling[8-9]. CLRFNet projects predicted charges onto a spatial grid and computes global electrostatic potential and electric field using the fast Fourier transform and an Ewald kernel. These methods show that explicit charge modeling can compensate for the limited description of long-range electrostatics in local potential models, although different methods treat charge constraints and energy consistency in different ways[2,15-17].

The charge equilibration (QEq) method starts from electronegativity equalization and represents atomic charges as the result of electronegativity, chemical hardness, and Coulomb interactions, with equilibrium charges solved under the total-charge constraint[7]. Compared with direct charge regression, QEq explicitly guarantees charge conservation. CE-ETNet combines this idea with an equivariant Transformer. Instead of directly predicting final charges, the model predicts atomic electronegativities from local environments, solves charge-balanced partial charges through a QEq linear system, and then computes the electrostatic energy[15-17]. In this way, the local equivariant representation describes the atomic environment, while QEq establishes charge equilibration and electrostatic coupling; together they form the final energy model.

3. Introduction to the CE-ETNet Model

This paper proposes CE-ETNet, an equivariant Transformer interatomic potential model based on charge-equilibration constraints[7,11,16]. It integrates local equivariant representations and charge-equilibration-based long-range Coulomb electrostatics within a unified computational framework for predicting potential energies and atomic forces of molecular systems, as shown in Fig. 1.

Figure 1: Schematic architecture of CE-ETNet



Note: (a) overall model structure; (b) equivariant representation encoder, which updates local atomic representations through multilayer interactions among distances, directions, and atomic features; (c) charge equilibration module, which constructs a constrained linear system from predicted electronegativities, interatomic Coulomb interactions, and total system charge to solve equilibrium partial charges and electrostatic energy; (d) output module, which receives atomic features and outputs the predicted energy and force.

For a system containing N atoms, the model takes as input the atomic numbers $z=(z_1, z_2,..., z_n)$, atomic coordinates $R=(r_1, r_2,..., r_n)$, and r_i in R_3. An adjacency graph is first constructed from atomic coordinates, and geometric edge features are generated from relative displacements, interatomic distances, and radial basis functions. The equivariant geometric representation encoder then learns local chemical environments through multilayer message passing and outputs atomic features containing both scalar and directional information. These features are fed into the short-range energy readout module and the charge equilibration module, respectively: the former produces the short-range energy E_{sr}, while the latter predicts atomic electronegativities, solves partial charges under the total-charge conservation constraint, and computes the electrostatic energy E_{ele}.

The total energy of the system is composed of the short-range energy and electrostatic energy:

$$E_{total} = E_{sr} + E_{ele} \tag{1}$$

The model does not introduce an independent force prediction head. Instead, it obtains atomic forces from the negative gradient of the total energy with respect to atomic coordinates. In addition, since the model uses relative positions and distances as geometric inputs, the total energy remains invariant under global translation or rotation, while the forces obtained from the energy gradient transform consistently with coordinate rotation. The following subsections describe the equivariant geometric representation encoder, differentiable charge equilibration module, and energy readout process.

3.1 Equivariant Geometric Feature Encoder

For atom i and a neighboring atom j, the relative displacement is defined as:

$$r_{ij} = r_j - r_i \tag{2}$$

The interatomic distance is:

$$d_{ij} = \left\| r_{ij} \right\| \tag{3}$$

In Eq. (1), r_i and r_j denote the spatial coordinates of atoms i and j, respectively; r_{ij} is the relative displacement vector from atom i to atom j, and d_{ij} is the Euclidean distance between the two atoms. A neighborhood is constructed using the cutoff radius r_c. When $d_{ij}<=r_c$, atom j is regarded as a neighbor of atom i[3-4].

The atomic number is then mapped to an initial scalar atomic feature:

$$h_i^{(0)} = Emb(z_i) \tag{4}$$

In Eq. (4), z_i is the atomic number of atom i, Emb(.) denotes a learnable element embedding function, and h_i^0 denotes the initial atomic feature. This feature mainly encodes atom type and basic chemical attributes, providing node representations for subsequent message passing.

To enable the network to distinguish atomic interactions at different distances, the interatomic distance is expanded into radial basis features:

$$\varphi(ij) = RBF\left(d(ij)\right) \tag{5}$$

In Eq. (5), RBF denotes radial basis function expansion, and the corresponding term represents the distance feature of the atom pair *(i,j)*. Compared with directly using a scalar distance, radial basis expansion describes interatomic distance variations at multiple scales, making it easier for the model to learn interaction patterns over different distance ranges[3-4].

On this basis, the model fuses the center-atom feature, neighbor-atom feature, and distance feature to generate an edge representation:

$$e_{ij}^{(l)} = f_e(h_i^{(l)}, h_j^{(l)}, g_{ij}) \tag{6}$$

In Eq. (6), the edge feature of atom pair *(i,j)* at layer l is obtained by a feature mapping function implemented with a multilayer perceptron, using the scalar features of atoms i and j at layer l. This step combines element information with geometric distance information and provides conditions for neighborhood message aggregation.

To emphasize neighboring atoms that contribute more strongly to the center atom, the encoder uses an attention mechanism to compute neighbor weights[11-12,14]:

$$\alpha_{ij}^{(l)} = \mathrm{softmax}_j\left(\frac{(q_i^{(l)})^{\mathrm{T}} k_{ij}^{(l)}}{\sqrt{d}}\right) \tag{7}$$

In Eq. (7), the attention weight measures the contribution of atom j when updating atom i. The query vector is mapped from the center-atom feature, the key vector is obtained by combining the neighbor-atom feature and edge feature, and d is the feature dimension. The softmax function is normalized over all neighbors of atom i, allowing the influence of different neighbors to be adaptively assigned.

Based on the attention weights, scalar information in the neighborhood is aggregated to obtain the local message of atom i:

$$m_i^{(l)} = \sum_{j \in \mathrm{N}(i)} \alpha_{ij}^{(l)} W_v e_{ij}^{(l)} \tag{8}$$

In Eq. (8), the neighborhood set of atom i is used to compute the aggregated local message through a learnable linear mapping. Through this process, atom i updates its chemical-environment representation according to local geometry and the types of neighboring atoms.

In addition to scalar features, the model constructs an equivariant update term using normalized relative displacement:

$$\Delta x_i^{(l)} = \sum_{j \in \mathrm{N}(i)} \beta_{ij}^{(l)} \frac{r_{ij}}{d_{ij}} \tag{9}$$

In Eq. (9), the update term represents the equivariant feature increment of atom *i*. The scalar coefficient is obtained from the edge-feature mapping and is independent of spatial rotation, while the unit direction vector of the atom pair rotates together with the coordinates. Therefore, this update process preserves rotational equivariance of the geometric features[5-6,10,13].

Finally, the local message and equivariant update are fused with the original feature, and the next-layer atomic feature is obtained through residual connection and normalization:

$$h_i^{(l+1)} = \mathrm{LayerNorm}\left(h_i^{(l)} + \mathrm{FFN}\left(\left[h_i^{(l)}, m_i^{(l)}, \Delta x_i^{(l)}\right]\right)\right) \tag{10}$$

In Eq. (10), the feed-forward neural network $FFN(\cdot)$ and feature concatenation produce the updated atomic scalar feature. After repeating the above process for *L* layers, the model obtains atomic features that contain atom types, local geometry, and directional relations, which are then fed into the short-range energy branch and the charge equilibration branch.

3.2 Differentiable Charge Equilibration Module

After the equivariant geometric feature encoder, each atom has an atomic representation containing local chemical-environment and geometric information. To further describe long-range electrostatic interactions, the network does not directly output atomic charges. Instead, it first predicts electronegativity parameters from atomic representations and then solves partial atomic charges using a charge equilibration equation with the total-charge constraint[7,15-17]. For atom *i*, the electronegativity is predicted as:

$$\chi_i = f_\chi\left(h_i^L\right) \tag{11}$$

In Eq. (11), h_i^L denotes the atomic scalar representation after *L* equivariant encoder layers, $f\chi(.)$ is the electronegativity prediction head, and χ_i denotes the effective electronegativity of atom *i* in the current chemical environment. This parameter is learned adaptively by the network and can therefore vary with local coordination, element type, and molecular conformation.

In the charge equilibration model, the electrostatic energy of the system is written as:

$$E_{\mathrm{QEq}}(q) = \sum_i \chi_i q_i + \frac{1}{2}\sum_i \sum_j q_i A_{ij} q_j \tag{12}$$

In Eq. (12), q_i denotes the partial charge of atom *i*, and A_{ij} is the matrix describing interatomic Coulomb interactions and atomic self-energy terms. The first term reflects the driving effect of electronegativity on charge distribution, while the second term represents Coulomb coupling among atomic charges and the hardness constraint. Compared with directly predicting q_i, this formulation converts charge assignment into a physically constrained energy minimization problem[7].

The diagonal elements of matrix *A* are given by atomic hardness, while the off-diagonal elements are constructed from Gaussian-smeared Coulomb interactions:

$$A_{ii} = J_i \tag{13}$$

$$A_{ij} = \frac{\mathrm{erf}\left(\frac{d_{ij}}{\sqrt{2}\gamma_{ij}}\right)}{d_{ij}}, \quad i \neq j \tag{14}$$

In Eq. (14), J_i denotes the atomic hardness parameter, which limits excessive charge concentration on a single atom; d_{ij} is the distance between atoms i and j; and gamma_i is determined by the atomic charge-width parameter, which avoids the singularity of the Coulomb term at short distances. The resulting A matrix contains both the self-charge cost of each atom and interatomic electrostatic interactions.

To ensure conservation of total system charge, the partial atomic charges must satisfy:

$$\sum_i q_i = Q_{\text{total}} \tag{15}$$

In Eq. (15), Q_{total} is the total charge of the input system.

The partial charges q and the Lagrange multiplier lambda are solved simultaneously using the following augmented linear equation:

$$\begin{bmatrix} A & \mathbf{1} \\ \mathbf{1}^T & 0 \end{bmatrix} \begin{bmatrix} q \\ \lambda \end{bmatrix} = \begin{bmatrix} -\chi \\ Q_{\text{total}} \end{bmatrix} \tag{16}$$

In Eq. (16), 1 is an all-one vector, and lambda is the Lagrange multiplier corresponding to the total-charge conservation constraint. The solution of this linear system is differentiable, so gradients from the energy and force losses can be back-propagated from the electrostatic energy to the electronegativity prediction head and the preceding equivariant representation encoder. Consequently, during training, the model can learn charge $d_i$stributions that are beneficial for energy and force prediction, rather than relying only on short-range information within a local cutoff neighborhood[15-17].

After the equilibrium charge q^* is obtained, the electrostatic energy is computed as:

$$E_{\text{ele}} = E_{\text{QEq}}\left(q^*\right) \tag{17}$$

In Eq. (17), q^* denotes the equilibrium partial charges solved under the total-charge constraint, and E_{ele} is the electrostatic energy output by the charge equilibration branch. This energy term is then combined with the output of the short-range potential branch for total-energy prediction.

3.3 Energy Readout and Force Prediction

The energy readout of CE-ETNet consists of a short-range potential branch and a long-range electrostatic branch. The short-range branch predicts the local energy contribution of each atom from the atomic representations output by the equivariant encoder, and sums over all atoms to obtain the short-range energy[1,5,11]:

$$E_{\text{sr}} = \sum_i \varepsilon_i\left(h_i^L, x_i^L\right) \tag{18}$$

In Eq. (18), ε_i denotes the short-range energy contribution of atom i, while h_i^L and x_i^L denote the scalar and equivariant features output by the encoder, respectively. This branch mainly describes short-range interactions determined by local chemical environments, such as near-neighbor bonding, repulsion, and the influence of local conformational changes on the potential energy.

The short-range energy is then added to the electrostatic energy obtained from the charge equilibration module to give the total energy of the system:

$$E_{\text{total}} = E_{\text{sr}} + E_{\text{ele}} \tag{19}$$

In Eq. (19), E_{sr} is the output of the short-range potential branch, and E_{ele} is the electrostatic energy obtained from the charge equilibration branch. Through this energy decomposition, the model retains the ability of equivariant neural networks to represent local geometric structure while explicitly introducing electrostatic contributions determined by partial charge distributions.

This work does not use a separate force prediction head. Instead, atomic forces are obtained as the negative gradient of the total energy with respect to atomic coordinates:

$$F_i = -\frac{\partial E_{\text{total}}}{\partial r_i} \tag{20}$$

This can be further expanded as:

$$F_i = -\frac{\partial E_{\text{sr}}}{\partial r_i} - \frac{\partial E_{\text{ele}}}{\partial r_i} \tag{21}$$

In Eq. (21), the first term represents the force contribution caused by the variation of the short-range potential with atomic coordinates, while the second term represents the force contribution caused by the electrostatic energy from charge equilibration. Since both force components are derived from the same total energy function, the model maintains consistency between energy and force prediction and avoids physical inconsistencies that may arise when energy and force are predicted separately.

During training, the model jointly optimizes the energy error and force error, with the loss function defined as:

$$L = \lambda_E L_E + \lambda_F L_F \tag{22}$$

In Eq. (22), L_E and L_F denote the mean absolute error or mean squared error losses for energy and force, respectively, and λ_E and L_F are the corresponding weights. Through joint energy-force training, the model improves the accuracy of atomic-force prediction while maintaining the continuity of the potential energy surface, making it more suitable for molecular dynamics simulations that require stability and physical consistency.

4. Experimental Analysis

4.1 QM9

To evaluate the effectiveness of CE-ETNet in molecular property prediction, experiments are first conducted on the QM9 dataset. QM9 contains about 130,000 small molecules with quantum-chemical properties and is widely used to assess the ability of models to learn structure-property relationships[18]. This paper selects 12 prediction targets, including polarizability, energy gap, HOMO, LUMO, dipole moment, heat capacity, free energy, enthalpy, electronic spatial extent, internal energy, zero-point corrected internal energy, and zero-point vibrational energy (ZPVE). CE-ETNet is compared with EQGAT, ET, HDGNN, Geoformer, Equiformer, and GotenNet[12,14]. The results are shown in Table 1. CE-ETNet achieves lower mean absolute errors on most prediction targets. These results indicate that, after introducing charge-equilibration constraints, the model can better describe properties related to molecular charge distributions and electrostatic interactions while retaining the ability to learn equivariant geometric features, thereby improving the overall performance on QM9 molecular property prediction.

Table 1:Mean Absolute Errors (MAE) and Units of Prediction Targets in QM9

Task	α	Δε	εHOMO	εLUMO	μ	Cv	G	H	R2	U	U0	ZPVE
Units	ma03	meV	meV	meV	mD	mcal	meV	meV	a02	meV	meV	meV
EQGAT	53	32	20	16	11	24	23	24	382	25	25	2.00
ET	59	36.1	20.3	17.5	11	26	7.62	6.16	33	6.38	6.15	1.84
HDGNN	46	32	18	16	17	23	11	10	342	8.12	8.34	1.21
Geoformer	40	33.8	18.4	15.4	10	22	6.13	4.39	28	4.41	4.43	1.28
Equiformer	46	30	15.4	14.7	12	23	7.63	6.63	251	6.74	6.59	1.26
Equiformerv2	47	29.0	14.4	13.3	9.9	23	7.57	6.22	186	6.49	6.17	1.47
GotenNet	33	21.2	16.9	13.9	7.5	20	5.50	3.70	29	3.67	3.71	1.09
CE-ETNet	31	20.3	15.3	12.9	7.7	19	5.35	3.55	26	3.50	3.57	1.09

4.2 Revised MD17

CE-ETNet is further evaluated on the revised MD17 dataset for molecular potential energy and atomic force prediction. Revised MD17 contains high-accuracy quantum-chemical energies and forces recalculated from the original MD17 molecular conformations, with approximately 100,000 configurations for each molecule[19]. In the experiments, 950 configurations are selected as the training set, 50 configurations as the validation set, and the remaining configurations as the test set for each molecule. CE-ETNet is compared with NequIP, ACE, BOTNet, MACE, TensorNet, and GotenNet[5,13-14]. The results are shown in Table 2. CE-ETNet obtains lower energy and force prediction errors for most molecular systems. Compared with the baseline GotenNet, CE-ETNet reduces both energy and force errors on Azobenzene, Ethanol, Malonaldehyde, Naphthalene, Paracetamol, Salicylic acid, Toluene, and Uracil. On Benzene, the energy error remains the same while the force error further decreases. Overall, after introducing the charge-equilibration electrostatic branch, CE-ETNet supplements local equivariant feature learning with charge-related electrostatic interactions, thereby improving energy and force prediction accuracy under the small-sample setting of revised MD17.

Table 2:Mean Absolute Errors (MAE) of Energies (kcalmol−1) and Forces (kcal mol−1Å−1) for 10 Small Organic Molecules in rMD17

Molecule	Aspirin		Azobenzene		Benzene		Ethanol		Malonaldehyde	
	Energy	Force	Energy	Force	Energy	Force	Energy	Force	Energy	Force
NequIP	0.0530	0.1891	0.0161	0.0669	0.0009	0.0069	0.0092	0.0646	0.0184	0.1176
ACE	0.1407	0.4128	0.0830	0.2514	0.0009	0.0115	0.0277	0.1683	0.0392	0.2560
BOTNet	0.0530	0.1960	0.0161	0.0761	0.0007	0.0069	0.0092	0.0738	0.0184	0.1338
MACE	0.0507	0.1522	0.0277	0.0692	0.0092	0.0069	0.0092	0.0484	0.0184	0.0945
TensorNet	0.0553	0.2052	0.0161	0.0715	0.0005	0.0069	0.0115	0.0807	0.0184	0.1245
GotenNet	0.0358	0.1306	0.0121	0.0483	0.0005	0.0047	0.0101	0.0482	0.0129	0.0830
CE-ETNet	0.0355	0.1342	0.0107	0.0461	0.0005	0.0045	0.0078	0.0469	0.0118	0.0795

Molecule	Naphthalene		Paracetamol		Salicylic acid		Toluene		Uracil	
	Energy	Force	Energy	Force	Energy	Force	Energy	Force	Energy	Force
NequIP	0.0208	0.0300	0.0323	0.1361	0.0161	0.0922	0.0069	0.0369	0.0092	0.0715
ACE	0.0208	0.1176	0.0922	0.2929	0.0415	0.2145	0.0254	0.1499	0.0254	0.1522
BOTNet	0.0046	0.0415	0.0300	0.1338	0.0184	0.0992	0.0092	0.0438	0.0092	0.0738
MACE	0.0115	0.0369	0.0300	0.1107	0.0208	0.0715	0.0115	0.0346	0.0115	0.0484
TensorNet	0.0046	0.0369	0.0300	0.1361	0.0184	0.1061	0.0069	0.0392	0.0092	0.0715
GotenNet	0.0045	0.0240	0.0244	0.0928	0.0141	0.0736	0.0044	0.0261	0.0064	0.0417
CE-ETNet	0.0039	0.0219	0.0212	0.0917	0.0128	0.0712	0.0039	0.0245	0.0060	0.0395

4.3 tmQM_wB97MV Dataset

CE-ETNet is also evaluated on the tmQM_wB97MV dataset to test its energy prediction ability for complex transition-metal complexes. The tmQM_wB97MV dataset was constructed by Garrison et al. from the structures of the tmQM dataset, with single-point energies recalculated using the higher-quality B97M-V/def2-SVPD electronic-structure level[20]. The original tmQM dataset is a large-scale quantum-chemical dataset of transition-metal complexes designed for homogeneous catalysis, covering 30 transition-metal elements such as Mn, Pd, and Ag. tmQM_wB97MV contains two subsets: tmQM_wB97MV with 86,507 structures and tmQM_wB97MV_neutral with 71,042 neutral structures. Compared with the small-molecule systems in QM9 and revised MD17, this dataset has more complex molecular compositions, with about 66 atoms per molecule on average, and therefore further tests the modeling ability of the method for large heterogeneous molecular systems. Following the original setting, the dataset is split into training, validation, and test sets with a ratio of 0.8, 0.1, and 0.1, and the mean absolute error (MAE) is used as the evaluation metric, with energy errors reported

in meV/atom. Table 3 compares CE-ETNet with SchNet, PaiNN, TensorNet, and GotenNet. On the full tmQM_wB97MV dataset, CE-ETNet achieves an energy MAE of 3.94 meV/atom, lower than 5.86 meV/atom of GotenNet. On the tmQM_wB97MV_neutral subset, CE-ETNet achieves an energy MAE of 3.22 meV/atom, also lower than 3.40 meV/atom of GotenNet. These results indicate that the electrostatic energy term introduced by the charge-equilibration constraint provides an effective supplement for transition-metal complexes, allowing the model to maintain good energy prediction accuracy under complex compositions and relatively large numbers of atoms.

Table 3 Energy MAE Comparison on tmQM_wB97MV and tmQM_wB97MV_neutral

Type		SchNet	PaiNN	TensorNet	GotenNet	CE-ETNet
tmQM_wB97MV	Energy	15.68	8.83	6.53	5.86	3.94
tmQM_wB97MV_neutral	Energy	7.60	5.76	3.81	3.40	3.22

5. Conclusion

Long-range electrostatic interactions have an important influence on the accurate prediction of molecular properties, interatomic potential energies, and atomic forces. This paper proposes CE-ETNet, an equivariant Transformer interatomic potential network based on charge-equilibration constraints. The model first uses an equivariant encoder to learn local geometric and elemental features of atoms, then predicts environment-dependent atomic electronegativities through an electronegativity head, and solves partial atomic charges and electrostatic energy under the total-charge conservation constraint using a differentiable QEq linear system. Finally, the electrostatic energy is added to the short-range potential energy to obtain the total energy, and atomic forces are computed consistently as the negative gradient of the total energy. The experimental results show that CE-ETNet outperforms baseline models on most molecular property prediction targets in QM9, further reduces energy and force prediction errors for most molecular systems compared with GotenNet on revised MD17, and achieves lower energy MAE on the tmQM_wB97MV transition-metal complex dataset. These results demonstrate that the charge-equilibration constraint effectively complements the description of electrostatic interactions in local equivariant models and improves prediction accuracy across different molecular systems. Future work will further consider long-range electrostatic treatment under periodic boundary conditions and validate the accuracy, stability, and computational efficiency of the model on larger material and interface systems.

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Conflicts of Interest

The authors declare no conflict of interest.

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