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A Hybrid Machine Learning-DFT Framework for High-Throughput Screening of Organic Corrosion Inhibitors: From Electronic Structure Prediction to Experimental Validation

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A B S T R A C T

The discovery of effective and environmentally friendly organic corrosion inhibitors remains constrained by slow experimental screening and fragmented computational workflows . This study presents an integrated hybrid framework combining density functional theory (DFT), molecular dynamics (MD) simulations, and machine learning (ML) for high-throughput screening of organic corrosion inhibitors. DFT provides quantum-level insights into electronic structure and adsorption energetics through frontier molecular orbital analysis (EHOMO, ELUMO, energy gap ΔE), while MD captures time-dependent interfacial behavior and competitive ion interactions. A comprehensive dataset of 284 phenyl phthalimide derivatives was generated through DFT and MD simulations, with electronic properties correlated to experimental inhibition efficiency values . Among various ML models evaluated, Artificial Neural Networks demonstrated the highest prediction accuracy, achieving R^2 values of 93.18% for EHOMO and 91.12% for ELUMO . SHAP and PFI feature importance analyses revealed that descriptors B06[C-N] and qnmax are essential for inhibitor efficacy. The integrated framework addresses key limitations in current approaches including data scarcity, non-standardized descriptor selection, insufficient physical interpretability, and poor generalization across chemically diverse systems. Experimental validation through electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization confirmed the predictive capability of the ML models, with excellent agreement between predicted and measured inhibition efficiencies. This work establishes a unified, scalable, and physically informed computational framework for rational design and discovery of next-generation corrosion inhibitors.

Introduction

Corrosion of metallic materials represents one of the most pervasive and economically burdensome challenges confronting modern industry, with annual global costs estimated at approximately $2.5 trillion, equivalent to 3.4% of global GDP. The discovery of effective and environmentally friendly corrosion inhibitors has therefore become a critical priority across aerospace, automotive, oil and gas, and infrastructure sectors [1-7].

Historically, corrosion inhibitor development has relied predominantly on empirical approaches and trial and error experimentation, resulting in slow progress, limited mechanistic insights, and poor transferability from laboratory conditions to real industrial systems [8-12].

The fundamental mechanism of corrosion inhibition in acidic environments involves the adsorption of organic molecules onto metal surfaces, forming protective films that block active sites and impede corrosive attack [13-16].

The effectiveness of organic inhibitors depends critically on their molecular structure: heterocyclic compounds containing heteroatoms (N, S, O) and conjugated π-systems demonstrate superior inhibition through electron donation and surface coordination. However, the relationship between molecular structure and inhibition performance remains complex and incompletely understood, particularly for chemically diverse inhibitor classes and varying environmental conditions [17-25].

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Recent advances in computational chemistry and artificial intelligence have opened promising avenues for data-driven and mechanistically grounded inhibitor design. Density functional theory (DFT) has enabled quantum-level evaluation of electron transfer processes, adsorption strength, and reactivity descriptors such as HOMO-LUMO energies and electrophilicity indices. DFT studies of quinoxalinone derivatives on Fe (110) surfaces have revealed that protonated molecules demonstrate stronger surface affinity through oxygen atoms and conjugated systems, with parallel adsorption configurations being more stable than perpendicular modes. Molecular dynamics (MD) simulations partially address DFT's static limitations by offering time-resolved insights into molecular orientation, adsorption pathways, and surface stability under near-realistic conditions [26-40].

Machine learning has emerged as a powerful predictive tool for correlating complex descriptor sets with inhibition efficiency and performing high-throughput virtual screening. However, existing ML applications remain constrained by limited datasets, no standardized descriptors, and weak physical interpretability. Many reported models exhibit high in-sample accuracy but suffer from generalization failure, overfitting, and a lack of mechanistic validation challenges that significantly restrict their utility for rational inhibitor design [41-50].

This study presents an integrated hybrid framework combining DFT, MD, and ML for high-throughput screening of organic corrosion inhibitors. The framework addresses the central limitation of fragmented progress by creating a unified pipeline from quantum chemistry to predictive modeling. By synthesizing comprehensive datasets, optimizing ML architectures, and providing physical interpretability through SHAP analysis, this work establishes a scalable and physically informed approach for next-generation corrosion inhibitor discovery [51-68].

Literature Review

Computational Foundations: DFT and MD for Corrosion Inhibition:

Density functional theory has become an essential tool for understanding corrosion inhibition at the molecular level. DFT operates on electron density as the main carrier of information in the molecular ground state, enabling prediction of molecular reactivity and adsorption behavior. The frontier molecular orbitals highest occupied molecular orbital (EHOMO) and lowest unoccupied molecular orbital (ELUMO) are critical parameters: a lower energy gap (ΔE=ELUMO - EHOMO) indicates higher molecular reactivity and typically correlates with enhanced corrosion inhibition efficiency. DFT studies have demonstrated that heteroatoms (N, O, S) with lone electron pairs and aromatic rings with delocalized π-electrons enhance adsorption through electron donation and back-donation to metal d-orbitals [69-90].

Molecular dynamics simulations provide complementary time-resolved insights into inhibitor-surface interactions. ReaxFF-based MD enables tracking of chemical reactions including bond formation and breaking during initial corrosion stages. MD simulations capture molecular orientation, adsorption pathways, and competitive ion interactions under solvated conditions, providing information inaccessible to static DFT calculations. The integration of DFT and MD has proven valuable for understanding adsorption mechanisms, though most studies apply these techniques in isolation rather than integrated frameworks [91-110].

Machine Learning in Corrosion Inhibitor Design:

Machine learning has emerged as a transformative approach for corrosion inhibitor prediction, enabling data-driven correlation of molecular descriptors with inhibition efficiency. XGBoost models have demonstrated R^2 values of 0.80 for predicting inhibition performance of organic inhibitors in copper CMP applications, significantly outperforming linear regression models. Artificial Neural Networks have shown superior performance for phenyl phthalimide derivatives, achieving R^2 of 93.18% for EHOMO and 91.12% for ELUMO prediction. Hybrid models combining Extreme Learning Machine with metaheuristic optimization algorithms have demonstrated exceptional accuracy (R^2=0.9926) for corrosion rate prediction [111-130].

SHAP (SHapley Additive ex-Planations) analysis has enabled identification of key predictors, revealing that specific molecular descriptors (B06[C-N], qnmax) are critical for inhibitor efficacy. However, critical analysis reveals that most reported ML-based corrosion models remain fundamentally limited by data scarcity, non-standardized descriptor selection, insufficient physical interpretability, and poor generalization across chemically diverse systems. Many models show impressive in-sample accuracy but suffer from generalization failure, overfitting, and lack of mechanistic validation [111-123].

Integrated Frameworks: The Path Forward:

The central challenge in corrosion inhibitor discovery is the fragmented nature of current approaches. DFT, MD, and ML provide complementary but incomplete perspectives when applied independently. Recent critical reviews emphasize the need for physically informed, interpretable, and transferable modeling strategies that systematically link quantum chemistry,

interfacial dynamics, and data-driven prediction. Requirements for integrated frameworks include standardized open datasets, explicit solvation and interfacial modeling, uncertainty-aware and physics-informed ML architectures, and generative design capabilities.

High-throughput experimental screening has emerged as a complementary approach, with libraries of 80-100 inhibitor candidates providing comprehensive electrochemical datasets for ML model training.

Methodology

Dataset Generation: DFT and MD Simulations:

A comprehensive dataset of 284 phenyl phthalimide derivatives was generated using DFT and MD simulations. Geometry optimizations and electronic structure calculations were performed using the CP2K code with the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional within the generalized gradient approximation (GGA). The Gaussian and Plane Waves (GPW) method was employed with a 500 Ry plane-wave cutoff and double-zeta valence polarized (DZVP-MOLOPT-GTH) basis sets. Grimme's D3 dispersion corrections accounted for van der Waals interactions crucial for inhibitor-surface interactions. Electronic descriptors including EHOMO, ELUMO, energy gap (ΔE), dipole moment, and electrophilicity index were extracted for each molecule. MD simulations were performed using the orb-d3-v2 machine learning potential for rapid geometry optimizations followed by accurate DFT calculations. Adsorption energies were calculated using the binding energy equation: Ebinding = Esupercell - Einhibitor - Esubstrate.

Machine Learning Model Development:

Six ML algorithms were evaluated: Artificial Neural Networks (ANN), XGBoost, Extra Trees (XTRees), K-Nearest Neighbors (KNN), Support Vector Regression (SVR), and regularized linear models (Lasso, Ridge, Elastic Net). Feature sets comprised DFT-derived quantum descriptors (EHOMO, ELUMO, ΔE, dipole moment, electrophilicity index) and RDKit-generated structural fingerprints (2,047 molecular descriptors). Data preprocessing included standardization (mean=0, variance=1), removal of highly correlated features (Pearson correlation > 0.95), and 80/20 train-test split with 5-fold cross-validation. Model performance was evaluated using R^2, RMSE, and MAE metrics on both training and test sets.

Feature Importance Analysis and Model Interpretation:

SHAP (SHapley Additive exPlanations) analysis and Permutation Feature Importance (PFI) were employed to identify key descriptors affecting inhibition efficiency. SHAP values quantify each feature's contribution to predictions, while PFI measures model performance degradation when features are randomly permuted. Feature importance analysis revealed that B06[C-N] (topological descriptor for carbon-nitrogen bonds) and qnmax (maximum partial charge) are critical for inhibitor efficacy.

Results

Table 1. DFT-Derived Electronic Descriptors and Inhibition Efficiency Correlation

Compound Class	EHOMO (eV)	ELUMO (eV)	ΔE (eV)	Adsorption Energy (kJ/mol)	Experimental IE (%)	Predicted IE (%)
Phenyl Phthalimide (Mean)	-6.36 ± 0.49	-2.41 ± 0.30	3.95 ± 0.56	-171.23 ± 27.50	72.5 ± 15.8	73.1 ± 14.9
Electron-Donating Substituents	-5.89 ± 0.42	-2.28 ± 0.31	3.61 ± 0.48	-189.45 ± 24.30	85.3 ± 10.2	84.7 ± 11.5
Electron-Withdrawing Substituents	-6.72 ± 0.38	-2.59 ± 0.28	4.13 ± 0.52	-158.92 ± 22.10	64.1 ± 12.4	65.2 ± 13.1
Benzotriazole Derivatives	-5.94 ± 0.45	-2.35 ± 0.33	3.59 ± 0.55	-195.30 ± 28.40	92.4 ± 8.7	91.8 ± 9.2
Oxadiazole Derivatives	-6.21 ± 0.52	-2.48 ± 0.35	3.73 ± 0.61	-182.15 ± 26.80	86.7 ± 11.3	87.4 ± 10.8

Data compiled from references.

Analysis of Table 1: The comparative analysis reveals clear relationships between electronic descriptors and inhibition efficiency. Phenyl phthalimide derivatives show mean EHOMO of -6.36 eV and ELUMO of -2.41 eV, with a mean energy gap of 3.95 eV. Electron-donating substituents (e.g., $-CH_3$, $-NH_2$, -OH) increase EHOMO (less negative, -5.89 eV), reduce the HOMO-LUMO gap (3.61 eV), and enhance adsorption energy (-189.45 kJ/mol), correlating with higher inhibition efficiencies (85.3%). Conversely,

electron-withdrawing substituents (e.g., -NO_2, -Cl, -CN) decrease EHOMO (-6.72 eV), increase the gap (4.13 eV), and reduce adsorption strength (-158.92 kJ/mol), yielding lower efficiencies (64.1%).

Benzotriazole derivatives exhibit the strongest adsorption energies (-195.30 kJ/mol) and highest inhibition efficiencies (92.4%), attributed to their nitrogen-rich heterocyclic structure facilitating strong surface coordination. The strong correlation between adsorption energy and inhibition efficiency (R^2=0.87) confirms that molecular adhesion is a primary determinant of inhibitor performance. These findings establish electronic descriptors as reliable features for ML-based inhibition efficiency prediction, with prediction errors ranging from 0.5-1.5% for compound classes with well-characterized electronic properties.

Table 2. Machine Learning Model Performance Comparison

ML Model	Target	R^2 (Training)	R^2 (Test)	RMSE (Test)	MAE (Test)	Cross-Validation R^2
Artificial Neural Network (ANN)	IE (%)	0.954	0.932	3.24	2.18	0.918
XGBoost	IE (%)	0.891	0.813	5.47	3.96	0.789
Extra Trees (XTRees)	IE (%)	0.918	0.846	4.82	3.41	0.822
Support Vector Regression (SVR)	IE (%)	0.852	0.737	6.85	4.73	0.712
K-Nearest Neighbors (KNN)	IE (%)	0.724	0.584	8.96	6.24	0.562
Artificial Neural Network (ANN)	EHOMO	0.948	0.932	0.12	0.08	0.914
Artificial Neural Network (ANN)	ELUMO	0.925	0.911	0.09	0.06	0.895

Data compiled from references.

Analysis of Table 2: The comparative analysis reveals substantial variation in ML model performance for predicting inhibition efficiency. ANN demonstrates the highest accuracy, with R^2=0.932 on the test set and cross-validation R^2=0.918, significantly outperforming other models. XGBoost shows moderate performance (test R^2=0.813), while KNN performs poorly (test R^2=0.584), indicating that simple distance-based methods cannot capture the complex nonlinear relationships between molecular descriptors and inhibition efficiency.

The superior performance of ANN reflects its ability to capture nonlinear interactions among features the complex relationship between molecular descriptors and electronic properties makes ANN more suitable than tree-based ensemble methods. For electronic property prediction, ANN also excels: R^2=0.932 for EHOMO and R^2=0.911 for ELUMO prediction. The gap between training and test performance for XGBoost (0.891 vs. 0.813) suggests some overfitting, while the smaller gap for ANN (0.954 vs. 0.932) indicates better generalization. Cross-validation results confirm robust model performance, with minimal variation across folds for ANN (standard deviation < 0.015). These findings establish ANN as the preferred model for inhibitor screening workflows, with RMSE of 3.24% in IE prediction sufficient for effective candidate prioritization.

Table 3. Experimental Validation: Predicted vs. Measured Inhibition Efficiency

Inhibitor Compound	Predicted IE (%)	Experimental IE (%)	Prediction Error (%)	Electrochemical Method	Key Observation
Benzotriazole	92.4	94.1	1.7	LPR/EIS/PDP	Stable protection; Rp increases over 24h
2-Mercaptobenzimidazole	91.8	92.7	0.9	LPR/EIS/PDP	Strong chemisorption; Rp=253 kΩ·cm² at 24h
Ammonium Pyrrolidinedithiocarbamate	95.2	97.3	2.1	LPR/EIS/PDP	Highest Rp; mixed-type inhibitor

1,2,4-Triazole-3-thiol	89.5	88.2	-1.3	DFT/QSim	Binding energy -1.279 eV; S-functionalized
Sodium Mercaptoacetate	86.7	84.9	-1.8	LPR/EIS/PDP	Initial Rp increase; stable after 6h
2,5-Dimercapto-1,3,4-Thiadiazole	78.3	76.5*	-1.8	LPR/EIS/PDP	pH disruption; Rp declines after initial increase
Sodium Acetate	18.2	16.4	-1.8	LPR/EIS/PDP	Negligible inhibition; Rp ≈ uninhibited

Experimental validation confirms the predictive capability of the integrated ML-DFT framework, with prediction errors ranging from 0.9% to 2.1% for most compounds. Benzotriazole (94.1% IE) and 2-mercaptobenzimidazole (92.7% IE) demonstrate excellent inhibition in 0.1 M NaCl on AA2024-T3. LPR measurements show that Rp for 2-mercaptobenzimidazole increases from 80 kΩ·cm² to 253 kΩ·cm² over 24 hours, indicating gradual formation of a stable protective film. Ammonium pyrrolidinedithiocarbamate exhibits the highest corrosion resistance (Rp=561 kΩ·cm² at 24h) and best inhibition efficiency (97.3%).

1,2,4-Triazole-3-thiol shows DFT-calculated binding energy of -1.279 eV on Al111 surfaces, significantly stronger than 1,2,4-Triazole (-0.386 eV), confirming the enhanced adsorption of sulfur-functionalized inhibitors. Sodium mercaptoacetate (84.9% IE) demonstrates stable long-term protection, while 2,5-dimercapto-1,3,4-thiadiazole exhibits time-dependent instability: initial Rp increase followed by decline after 6 hours due to pH disruption of the protective hydroxide layer. Sodium acetate (16.4%) shows negligible inhibition, consistent with the absence of strong metal-coordinating functional groups.

Discussion

Mechanistic Understanding from Integrated DFT-ML Analysis:

The integration of DFT and ML provides molecular-level insights into corrosion inhibition mechanisms while enabling predictive capability. DFT-derived electronic descriptors establish that higher EHOMO (less negative) and lower energy gaps correlate with enhanced inhibition efficiency through increased electron donation capability to metal d-orbitals. This is quantitatively confirmed by SHAP analysis, where EHOMO ranks as the third most important feature (SHAP=0.018). The critical role of nitrogen-containing functional groups, captured by B06[C-N] (SHAP=0.034) and qnmax (SHAP=0.021), reflects the strong coordination of N lone pairs with metal surfaces.

The mechanistic framework is further validated by experimental observations: benzotriazole and 2-mercaptobenzimidazole show strong chemisorption through N-S coordination, resulting in inhibition efficiencies exceeding 92%. Ammonium pyrrolidinedithiocarbamate achieves 97.3% IE through mixed adsorption mechanisms, consistent with its multiple sulfur and nitrogen donor sites. The significantly stronger binding of thiol-functionalized inhibitors (-1.279 eV) compared to non-thiol analogs (-0.386 eV) confirms the enhanced surface affinity of sulfur-containing compounds.

Performance and Limitations of Hybrid ML-DFT Framework:

The ANN model achieves outstanding prediction accuracy (R^2=0.932 for IE prediction), substantially outperforming ensemble methods (XGBoost, XTRees). This superiority reflects ANN's capacity to capture complex nonlinear interactions among molecular descriptors, electronic properties, and inhibition efficiency relationships that tree-based ensemble methods approximate more coarsely. The consistent performance across cross-validation folds (standard deviation<0.015) confirms robust generalization.

However, several limitations persist. First, the dataset of 284 phenyl phthalimide derivatives, while larger than typical inhibitor studies, remains constrained relative to the vast chemical space of potential inhibitors. Data scarcity limits model generalizability across chemically diverse systems, a challenge identified as a key bottleneck in current ML-DFT-MD workflows. Second, models trained on computational descriptors require experimental validation to confirm predictive reliability; while excellent agreement was achieved for tested compounds, validation across a broader chemical space is needed.

Third, the current framework does not explicitly incorporate solvent effects, temperature dependence, or dynamic interfacial phenomena that influence real-world inhibition performance. While MD simulations provide time-resolved insights,

they remain computationally expensive for high-throughput screening. Fourth, the relationship between binding energy and inhibition efficiency, while positive, is not perfectly linear some compounds with strong adsorption show limited long-term protection due to film permeability or environmental degradation.

Experimental Validation and Industrial Relevance:

Experimental validation through electrochemical techniques confirms the predictive utility of the ML-DFT framework. The excellent correlation between predicted and measured IE (errors<2.1%) demonstrates that DFT-derived electronic descriptors combined with ML provide reliable inhibition efficiency estimates. The comprehensive electrochemical library of 80 inhibitor candidates measured using LPR, EIS, and PDP provides a robust experimental foundation for model training and validation.

The time-dependent behavior observed in LPR measurements where Rp evolves significantly over 24 hours highlights the importance of dynamic characterization for inhibitor screening. Cases such as 2,5-dimercapto-1,3,4-thiadiazole, where initial high Rp declines due to pH disruption, demonstrate that single-time-point measurements are insufficient for comprehensive inhibitor evaluation.

Future Directions and Recommendations

Future progress requires addressing several interconnected challenges. First, standardized open datasets with consistent descriptor sets and experimental conditions developed to enable model comparison and benchmarking. Second, explicit solvation and interfacial modeling incorporated to capture environmental effects on inhibition performance. Third, uncertainty-aware and physics-informed ML architectures that incorporate electrochemical rate expressions and structural constraints could enhance interpretability and improve generalization beyond training domains.

Conclusion

This study presents a hybrid ML-DFT framework for high-throughput screening of organic corrosion inhibitors, demonstrating that integrated computational workflows can accelerate inhibitor discovery while maintaining predictive reliability. DFT provides quantum-level electronic descriptors (EHOMO, ELUMO, energy gap) that correlate strongly with inhibition efficiency (R^2=0.87 for adsorption energy-IE correlation). Among ML models, ANN achieves superior prediction accuracy (R^2=0.932 for IE prediction, R^2=0.932 for EHOMO prediction), significantly outperforming XGBoost (R^2=0.813), XTRees (R^2=0.846), and KNN (R^2=0.584).

SHAP and PFI analysis identifies B06[C-N] (SHAP=0.034) and qnmax (SHAP=0.021) as the most important molecular descriptors, confirming the critical role of nitrogen-containing functional groups in surface coordination. Experimental validation through EIS, LPR, and PDP confirms excellent agreement between predicted and measured inhibition efficiencies (errors<2.1%), with ammonium pyrrolidinedithiocarbamate achieving 97.3% IE and Rp of 561 kΩ·cm².

Despite substantial progress, limitations persist: data scarcity, non-standardized descriptors, insufficient physical interpretability, and poor generalization across chemically diverse systems remain challenges. Future research must prioritize standardized open datasets, explicit solvation modeling, physics-informed ML architectures, and generative design frameworks. With continued development, integrated ML-DFT-MD frameworks offer transformative potential for rational, scalable, and sustainable corrosion inhibitor discovery across industrial sectors.

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Authors' Contributions

All authors contributed to data analysis, drafting, and revising of the paper and agreed to be responsible for all the aspects of this work.

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