

BenchQC: A Benchmarking Toolkit for Quantum Computation

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Abstract

The Variational Quantum Eigensolver (VQE) is a widely studied hybrid classical-quantum algorithm for approximating ground-state energies in molecular and materials systems. This study benchmarks the performance of the VQE for calculating ground-state energies of small aluminum clusters (Al^- , Al_2 , and Al_3^-) within a quantum-density functional theory (DFT) embedding framework, systematically varying key parameters — (I) classical optimizers, (II) circuit types, (III) number of repetitions, (IV) simulator types, (V) basis sets, and (VI) noise models. All calculations were performed using quantum simulators to evaluate VQE performance under both idealized and noise-augmented conditions. Our findings demonstrate that certain optimizers converge efficiently, while circuit choice and basis set selection have a marked impact on energy estimates, with higher-level basis sets closely matching classical computation

data from Numerical Python Solver (NumPy) and Computational Chemistry Comparison and Benchmark DataBase (CCCBDB). To approximate realistic conditions, we employed IBM noise models to simulate the effects of hardware noise. The results showed close agreement with CCCBDB benchmarks, with percent errors consistently below 0.2 %. The results demonstrate that VQE can approximate energy estimates under simulated conditions for small aluminum clusters and highlight the importance of optimizing quantum-DFT parameters to balance computational cost and precision. This work contributes to ongoing efforts to benchmark VQE in practical settings and lays the groundwork for future benchmarking tools for quantum chemistry and materials applications.

Introduction

Quantum computing represents a shift in computational technology, leveraging the principles of quantum mechanics to process information in ways that classical computers cannot.^{1,2} Unlike classical bits, which are binary and represent either 0 or 1, quantum bits, or qubits, can exist in a superposition of states, enabling them to perform many calculations simultaneously. This parallelism, combined with phenomena such as entanglement and quantum interference, gives quantum computers the potential to solve certain problems exponentially faster than their classical counterparts.^{3,4} Because of their potential to solve complex problems, quantum computers are at the forefront of innovation in fields like chemistry and materials science.

The need for quantum computing in chemistry and materials discovery stems from the complexity of molecular systems and the vast number of configurations that must be explored to identify new materials.⁵⁻⁷ Traditional computational methods, such as density functional theory (DFT) and post-Hartree-Fock approaches, provide valuable insights but often fall short when applied to large systems and strongly correlated electrons, or when high accuracy is required.⁸⁻¹¹ Materials discovery is a field in which the identification of new compounds with desired properties, such as high-temperature superconductivity, enhanced

catalytic activity, or improved energy storage, can revolutionize industries. However, the challenge lies in accurately predicting the properties of complex materials before they are synthesized.¹² Quantum computing offers a promising long-term avenue to address these challenges by enabling the precise simulation of quantum systems, allowing researchers to explore the electronic structure and properties of materials at an unprecedented level of detail. Although quantum computing has the potential to revolutionize chemistry and materials science, current noisy intermediate-scale quantum (NISQ) devices face significant limitations. These devices are constrained by noise and limited qubit counts, restricting the size of systems that can be effectively simulated using solely quantum methods.^{2,13} To address these challenges, quantum-DFT embedding integrates classical and quantum computing approaches, offering a potential solution that mitigates the hardware constraints of NISQ devices.^{7,14–16}

This hybrid strategy leverages DFT for less correlated electrons (core electrons) and quantum computing for the strongly correlated region (valence electrons), enabling simulations of systems beyond what current NISQ devices can handle alone. One of the key challenges in quantum chemistry is accurately capturing the behavior of strongly correlated electrons, particularly in materials with complex electronic structures. Quantum algorithms such as the Variational Quantum Eigensolver (VQE) play a crucial role in the quantum region of these simulations.^{11,17–19} The VQE is particularly well-suited for use with NISQ devices because of its hybrid nature.¹⁷ It utilizes a classical optimizer to minimize the energy of a quantum system, represented as a parameterized quantum circuit.²⁰ By iterating between quantum measurements and classical optimization, the VQE can approximate the ground-state energy of complex systems, providing a path to more accurate and efficient simulations of molecular and material properties.^{19,21} The integration of VQE into the quantum-DFT embedding framework enables researchers to tackle challenging problems in chemistry and materials science. For example, it can help explore the electronic structure of systems with strongly correlated electrons, such as transition-metal complexes.²² This approach offers an

additional route to achieving the high precision needed for materials discovery while still mitigating the limitations of current quantum hardware. As quantum computers continue to advance, the VQE, alongside quantum-DFT embedding, could potentially enhance the predictive capabilities in chemistry and materials science, offering new insights into phenomena that were previously beyond reach.

To date, the VQE algorithm has been employed to analyze the electronic structure of smaller molecular systems such as H_2 , LiH , and BeH_2 .^{6,17,20,23–25} It has also been applied to simulate electron and phonon band structures for materials applications.²⁶ While prior studies apply the VQE to simple molecules, systematic benchmarking frameworks tailored for quantum-DFT embedding are lacking.²⁷ Other recent works have focused on improving VQE performance through machine learning-based initial point selection, demonstrating enhanced convergence efficiency and reduced risk of getting trapped in local minima.^{28–30} In this work, we evaluate the performance of VQE for simulations of small aluminum clusters using a previously developed quantum-DFT embedding workflow by Pollard et al.³¹ All quantum calculations performed in this study are carried out using quantum simulators. Although these simulations do not capture the full complexity of hardware execution, they allow for controlled benchmarking of algorithm behavior and noise resilience. Our results show that the VQE can produce low-energy estimates under simulated conditions when integrated into an embedding framework. Unlike previous studies that focus on proposing new algorithms or specific system demonstrations, this work emphasizes reproducibility and systematic parameter exploration. Our contribution lies in presenting a reusable benchmarking framework tailored for quantum–DFT embedding, which can be extended for broader studies as quantum hardware continues to evolve. This study represents a step toward expanding the applicability of the VQE, demonstrating its potential for use in chemical simulations under NISQ-era constraints. Furthermore, our findings highlight the effectiveness of combining VQE with quantum-DFT embedding, a hybrid method that mitigates the limitations of current quantum devices, offering a resource-efficient approach for studying targeted quantum

subsystems within complex materials.

Methodology and Computational Details

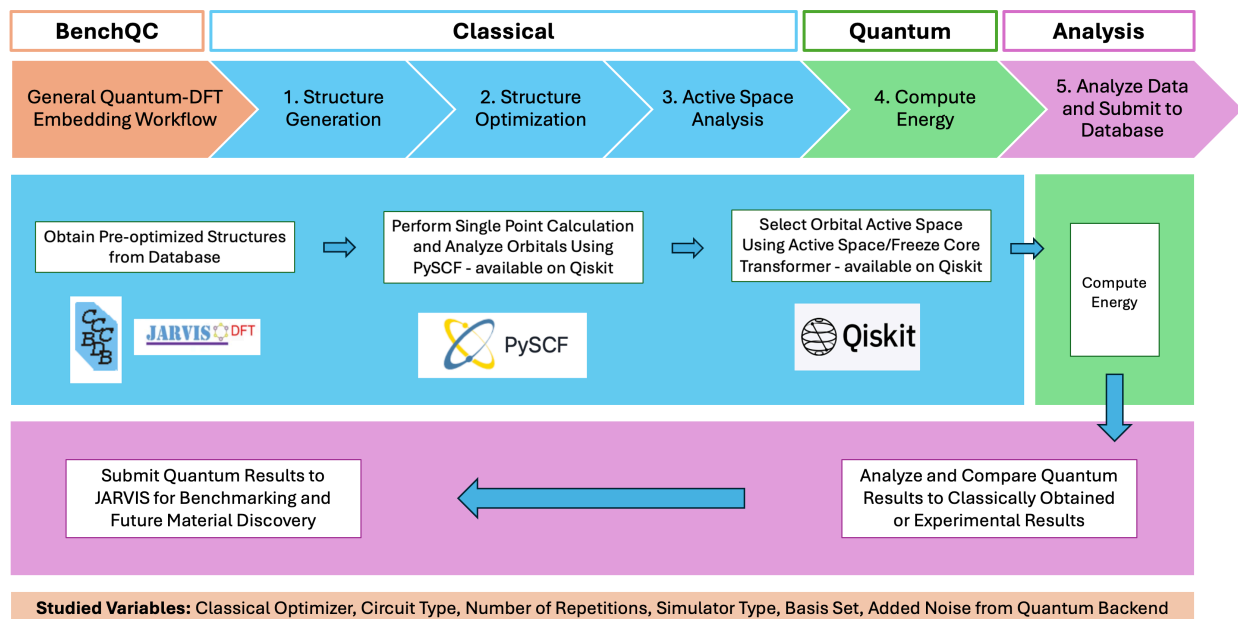


Figure 1: General Quantum-DFT Embedding Workflow: The workflow begins with structure generation where pre-optimized structures are obtained from CCCBDB, JARVIS-DFT, or self-generated from previous studies. Single point calculations are then performed to analyze the orbital active space. Once the active space is determined, the energy can be computed using either the quantum simulator or hardware. Results are then analyzed and compared to classically obtained or experimental results.

The quantum-DFT embedding workflow utilized in this study was employed on IBM's open-source platform for quantum computing, Qiskit³² (Version 43.1). The workflow consists of five main steps as shown in Figure 1. The first step is structure generation. Pre-optimized

structures are obtained from external databases, such as the Computational Chemistry Comparison and Benchmark Database (CCCBDB)³³ and the Joint Automated Repository for Various Integrated Simulations (JARVIS-DFT).^{34,35} These databases provide the necessary starting geometries for subsequent simulations. Structures can also be obtained from previous work or self-generated in any molecular visualization software (i.e. Avogadro^{36,37}). After structure generation, the PySCF package³⁸⁻⁴⁰ (available within the Qiskit framework) is utilized to perform single-point calculations on the pre-optimized structures. This step analyzes molecular orbitals to prepare for the selection of the active space in the next phase of the workflow. The appropriate orbital active space is then determined using the Active Space Transformer available on Qiskit Nature.⁴¹ This step is crucial for focusing the quantum computation on the most important part of the system, ensuring computational efficiency without sacrificing accuracy. The quantum region, consisting of the selected active space, is then passed to a quantum simulator or quantum hardware. The quantum computation is performed to calculate the energy of the system. Once the quantum computation is completed, the results are analyzed and compared to data obtained from Numerical Python (NumPy) or experimental results. NumPy is used as a reference for benchmarking VQE results because it performs exact diagonalization of the Hamiltonian within the defined active space and basis set. This approach yields precise ground-state energies free from noise or approximations beyond the chosen basis set, making these values reliable classical benchmarks for evaluating the accuracy of the VQE algorithm. The quantum simulation results are then submitted to the JARVIS leaderboard for benchmarking and further use in material discovery or design efforts. This integrated workflow leverages classical and quantum resources, enabling the simulation of complex materials by combining the strengths of both computational models.

In this work, we utilized both self-generated and pre-optimized aluminum molecules (ranging from Al^- to Al_3^-) (Figure 2) from a previous study.³¹ Note that all systems with an odd number of electrons were assigned an additional negative charge. This adjustment is necessary due to a limitation of the developed workflow³¹ and the `ActiveSpaceTransformer`

from Qiskit Version 43.1, which requires both the active and inactive spaces to contain an even number of electrons; therefore, there cannot be any unpaired electrons. Further updates to Qiskit past version 43.1 may enable calculations for systems with unpaired electrons. Aluminum clusters were chosen for their intermediate complexity and significant relevance to materials science, particularly in applications like catalysis, etc.^{42–44} Their well-characterized properties and the availability of reliable classical benchmarks, such as those from the CC-CBDB, make them ideal systems for validating quantum algorithms and exploring potential applications in materials discovery. Single-point energy calculations were performed on pre-optimized aluminum structures using the open-source software package PySCF, which is integrated as a driver within the Qiskit interface. We employed the default functional, local density approximation (LDA),⁴⁵ as available in Qiskit Version 43.1, and varied several key parameters, including the basis set, the classical optimizer, the type of circuit, the number of repetitions (reps), and quantum simulator type. For consistency, the default parameters included a STO-3G basis set, the Sequential Least Squares Programming (SLSQP) optimizer, an EfficientSU2 ansatz with 1 repetition, and the Statevector simulator. These choices reflect commonly used settings for VQE calculations and serve as a baseline for parameter optimization. We note that the hardware-efficient EfficientSU2 ansatz is composed of single-qubit rotation gates and entangling layers. It is widely adopted in NISQ-era algorithms due to its compatibility with hardware constraints and tunable expressiveness via repetition. While it does offer practical advantages, it does not conserve physical symmetries such as particle number or spin which can limit accuracy in some systems. More chemically informed ansatzes, such as the unitary coupled cluster (UCC) family, better preserve these symmetries and may offer improved performance in strongly correlated systems. However, such ansatzes generally require deeper circuits and more gates, making them less feasible on current quantum hardware. This study therefore focuses on a trade-off: evaluating simulation performance using a low-depth ansatz under realistic noise and resource constraints.

Following the initial single-point calculation, an active space of three orbitals (two filled

and one unfilled) or four electrons was selected. The choice of active space is based on a previous study, which demonstrated that this configuration provided accurate results for smaller aluminum clusters.³¹ The reduced Hamiltonian was subsequently printed, and the quantum states were encoded into qubits via Jordan Wigner Mapping. Each quantum simulation used 6 qubits, corresponding to 3 spatial orbitals mapped to spin orbitals using the Jordan–Wigner transformation (2 qubits per spatial orbital). This linear mapping of orbitals to qubits is standard for fermionic Hamiltonians and ensures that the circuit size scales proportionally with the size of the active space. Initial calculations were performed on quantum simulators with plans to later replicate on quantum hardware to determine the consistency of results. Due to limitations, no actual quantum hardware was utilized for this study.

Results and discussion

I. Analyzing electronic structure with a quantum computer

In this study, we employ a quantum-DFT embedding framework to benchmark the performance of the VQE for chemical problems and analyze the electronic structure of the studied aluminum systems. Figure 2 presents the visualization of the electronic density of the highest occupied molecular orbitals (HOMO) and lowest unoccupied molecular orbitals (LUMO) for Al^- , Al_2 , and Al_3^- . These visualizations were obtained using the program VESTA.⁴⁶ Each system is rotated (as indicated by the accompanying compass) to provide a clearer view of the electron densities. The most interesting result of these visualizations is the LUMO of Al_3^- , where notable electron delocalization is observed. We hypothesize that this delocalization is induced by the additional negative charge, which likely increases electron-electron repulsion within the system, thereby affecting the spatial distribution of the electrons. This behavior is consistent with the trends seen in other anionic systems, where excess negative charge can influence molecular orbitals and the distribution of electron density.⁴⁷

Figure 3 shows a graph of the ground-state energy vs. bond distance for Al_2 , where we

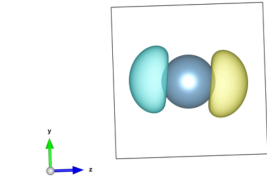
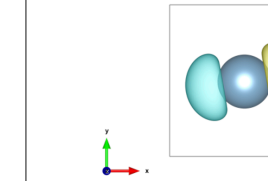
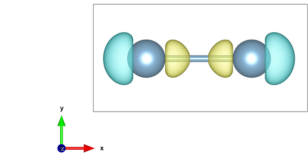
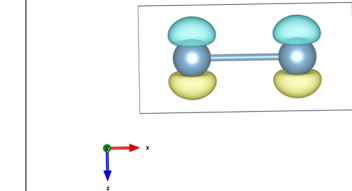
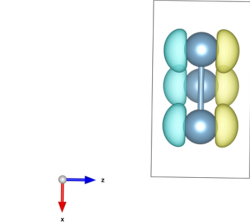
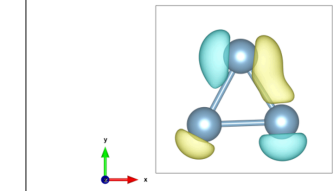
	Highest Occupied Molecular Orbital	Lowest Unoccupied Molecular Orbital
Al^-		
Al_2		
Al_3^-		

Figure 2: Visualization of the electron density of the highest occupied molecular orbitals (HOMOs) and the lowest unoccupied molecular orbitals (LUMOs) of the studied systems with an included compass. Systems were rotated to better visualize the electron density. Results obtained using the default parameters: LDA functional, a STO-3G basis set, SLSQP optimizer, an EfficientSU2 ansatz with 1 repetition, and the Statevector simulator.

compute the ground-state energy at various interatomic distances. The shape of the plot is consistent with what is typically expected for a diatomic molecule such as Al_2 .⁴⁸ The energy drops sharply as the atoms approach each other, reaching a minimum at the equilibrium bond distance. This minimum corresponds to the most stable configuration of the molecule. Beyond this point, the energy remains relatively flat then begins to increase slightly as the bond distance continues to increase, reflecting the dissociation limit where the atoms are no longer interacting strongly. The equilibrium bond distance of Al_2 is typically around 0.25 nm - 0.27 nm ($2.5 \text{ \AA} - 2.7 \text{ \AA}$),⁴⁹ which is consistent with the region where the plot reaches a minimum in our calculations.

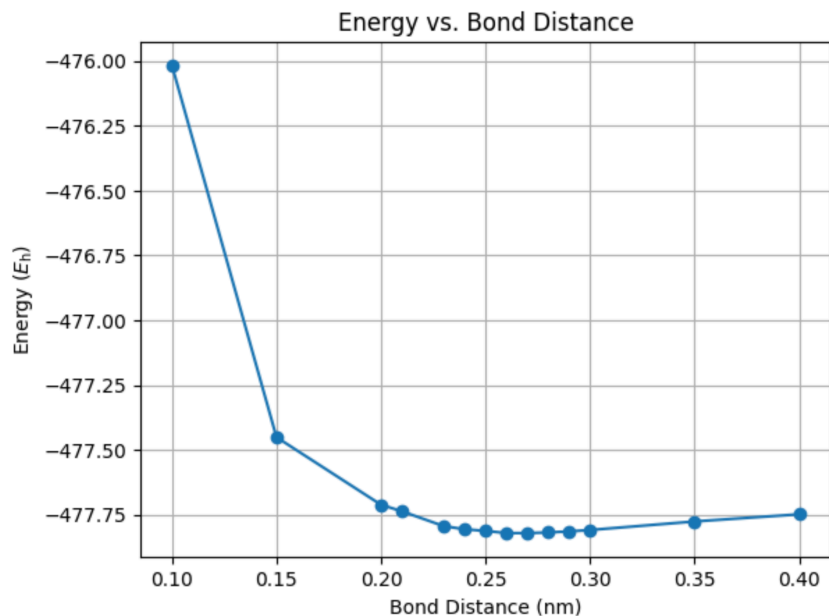


Figure 3: Ground-State Energy of Al_2 at Various Interatomic Distances: The graph begins to converge at around 0.25 nm - 0.27 nm ($2.5 \text{ \AA} - 2.7 \text{ \AA}$), which is consistent with the typical equilibrium bond distance of Al_2 . VQE energy results obtained using the default parameters: LDA functional, a STO-3G basis set, SLSQP optimizer, an EfficientSU2 ansatz with 1 repetition, and the Statevector simulator.

II. Benchmarking the performance of the VQE algorithm for chemistry problems

To benchmark the performance of the VQE algorithm for estimating ground-state energy values of the small aluminum clusters, we systematically varied several key parameters, including classical optimizers, circuit type, number of reps, simulator type, and basis set. As reference values for benchmarking the VQE results, we employed the Python library NumPy for exact diagonalization of the Hamiltonian within the defined active space and basis set. We compared the results from both NumPy and our quantum calculations using different basis sets with classically obtained data from the CCCBDB. All energies are reported in Hartree (E_h) (see conversion to Joules used in the International System of Units¹), as it is the natural unit in quantum mechanics and widely used in quantum chemistry to ensure consistency, particularly for benchmarking studies.

The first parameter we analyzed was the classical optimizer. In quantum-DFT embedding, classical optimizers are used to adjust parameters in hybrid quantum-classical algorithms, like VQE, to minimize the energy of the quantum region by optimizing the interaction between the quantum and classical parts of the system.⁵⁰ This ensures accurate and efficient calculations of strongly correlated regions within the framework. The optimizers we tested included Sequential Least Squares Programming (SLSQP), Limited-memory Broyden-Fletcher-Goldfarb-Shanno Bound (L_BFGS_B), Constrained Optimization By Linear Approximations (COBYLA), and Simultaneous Perturbation Stochastic Approximation (SPSA), with SLSQP serving as the default optimizer for all other calculations. SLSQP minimizes a scalar function subject to bounds, equality, and inequality constraints using gradient-based methods. It converges relatively quickly with stable performance in energy minimization tasks. L_BFGS_B is an optimization algorithm that approximates the Broyden-Fletcher-Goldfarb-Shanno (BFGS) method, designed to handle large-scale problems with bound constraints but is often slower to converge. COBYLA is an iterative, non-

¹Conversion of Hartree Energy to SI Unit Joules: $1 E_h = 27.2114 \text{ eV} = 4.3597\text{e-}18 \text{ J}$

linear, derivative-free constrained optimization algorithm that uses a linear approximation approach. The algorithm is easy to use for a small number of variables. SPSA is a derivative-free optimization method that efficiently estimates gradients through random perturbations, making it well-suited for noisy functions.^{26,51}

Our analysis of the different optimizers revealed that the VQE energy values differed slightly, with COBYLA and SPSA producing energy values slightly higher than those obtained with SLSQP and L_BFGS_B, while the NumPy values remained unchanged, as expected (Table 1). Figure 4 presents a visual representation of the performance of each optimizer for the calculation of Al_2 . This analysis highlights that the COBYLA optimizer is the most efficient, converging the fastest in comparison to the other three optimizers. SLSQP performs similarly but takes a bit longer to stabilize. SPSA initially converges more slowly but stabilizes earlier than L_BFGS_B, which proved to be the least efficient, requiring the most steps to converge and showing notable fluctuations throughout the evaluation process. Notably, this trend was consistent across all three studied systems. These results agree with studies on other aluminum-based systems.²⁶

The data tables (Table 1) further support these observations, showing that all optimizers yield VQE energy values close to the NumPy reference values, differing by at most 0.0118%. The variation (\pm) column reports the maximum observed deviation in VQE energy results across multiple runs with identical parameters. This reflects non-deterministic behavior, which can be due to optimizer initialization or sampling noise in the simulation backend. Each configuration was repeated five times to capture this variability. SLSQP and L_BFGS_B produce the most consistent and accurate results with minimal to no deviation across all systems. This aligns with the convergence graph analysis, where SLSQP converged smoothly and L_BFGS_B, although slightly more erratic, still provided accurate results. The near-zero deviation also highlights the stability of these optimizers. COBYLA shows slightly more variability, with minor deviations (e.g., 0.000 037 for Al^- and 0.000 022 for Al_2). Although these deviations are small, they suggest that COBYLA may introduce

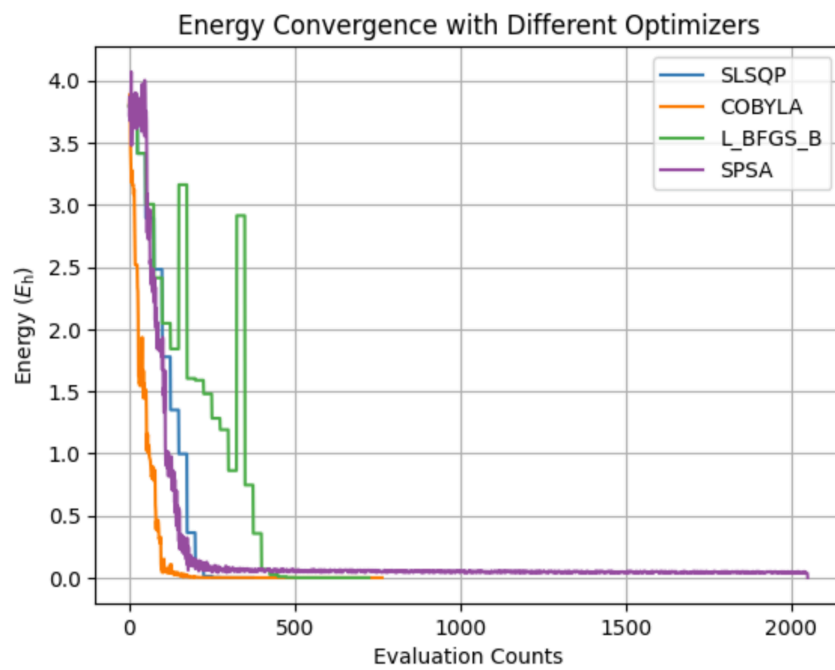


Figure 4: Energy Convergence of VQE with Varied Optimizers Rendered Using Qiskit.³² The COBYLA optimizer appears to converge the fastest in comparison to SLSQP, L_BFGS_B and SPSA.

minor fluctuations, which we also observed in the convergence graph where it converged quickly but with slight variations. Interestingly, despite its faster convergence in terms of energy values, COBYLA required longer job times compared to SLSQP (e.g., 16.4 seconds vs. 14.1 seconds for Al⁻) (Table S1). This discrepancy is likely due to COBYLA’s reliance on more function evaluations per iteration. SPSA produces results that are close to the reference values but shows a higher energy values for Al₂ and (Table 1b). This is consistent with the convergence graph, where SPSA initially converged more slowly and took longer to stabilize. SPSA is generally more robust in noisy conditions, but it may not be as accurate or efficient for ideal simulations. This alignment across both the convergence graph and tables indicates that SLSQP is the best choice for accuracy and stability, while COBYLA and SPSA may be preferable for applications where quick convergence or tolerance to noise is more critical.

The next parameter we analyzed was the circuit type or the ansatz. The selection of quantum circuit models is largely intuitive and dependent upon the specific problem being addressed.^{26,52} We utilized six different ansatzes for the simulation of Al⁻, Al₂, Al₃⁻, with all other parameters being held constant. An ansatz is a parameterized quantum circuit designed to approximate a quantum state.^{17,18} The ansatz is a hypothesis about the form of the wavefunction and is optimized to minimize the energy of the system. All six circuit models discussed here are readily available in JARVIS-tools, specifically within the *Jarvis.core.circuits.QuantumCircuitLibrary* module.³⁴ These circuits can all be visualized in Figure 5. Circuits 4, 5, and 6 are also known as RealAmplitudes, PauliTwoDesign and EfficientSU2 circuits, respectively.

We observed that circuits #1, #2, #3, #4, and #6 provide predictions of the ground-state energy estimates with less than 0.01 % difference from the NumPy reference values (Table 2). These circuits also exhibit near-identical VQE energies with negligible deviations across the systems, suggesting these circuits are stable and reliable for these simulations. In contrast, Circuit #5 (PauliTwoDesign) consistently shows a larger deviation (approximately

Table 1: Comparison of Ground-State Energy (E_h) when Varying Classical Optimizers (SLSQP, L_BFGS_B, COBYLA, SPSA) on Statevector Simulator, using Default Parameters: STO-3G Basis Set, Efficient SU2 Ansatz (Circuit #6), and 1 Repetition

(a) Al^-

	Al^- (VQE)	Variation (\pm)	Al^- (NumPy)
SLSQP	-238.842 31	0.000 000	-238.870 47
L_BFGS_B	-238.842 29	0.000 000	-238.870 47
COBYLA	-238.843 67	0.000 037	-238.870 47
SPSA	-238.843 47	0.000 000	-238.870 47

(b) Al_2

	Al_2 (VQE)	Variation (\pm)	Al_2 (NumPy)
SLSQP	-477.799 05	0.000 000	-477.801 97
L_BFGS_B	-477.799 05	0.000 000	-477.801 97
COBYLA	-477.799 03	0.000 022	-477.801 97
SPSA	-477.793 62	0.000 000	-477.801 97

(c) Al_3^-

	Al_3^- (VQE)	Variation (\pm)	Al_3^- (NumPy)
SLSQP	-716.675 25	0.000 000	-716.708 73
L_BFGS_B	-716.679 28	0.000 031	-716.708 73
COBYLA	-716.690 60	0.000 000	-716.708 73
SPSA	-716.698 41	0.000 000	-716.708 73

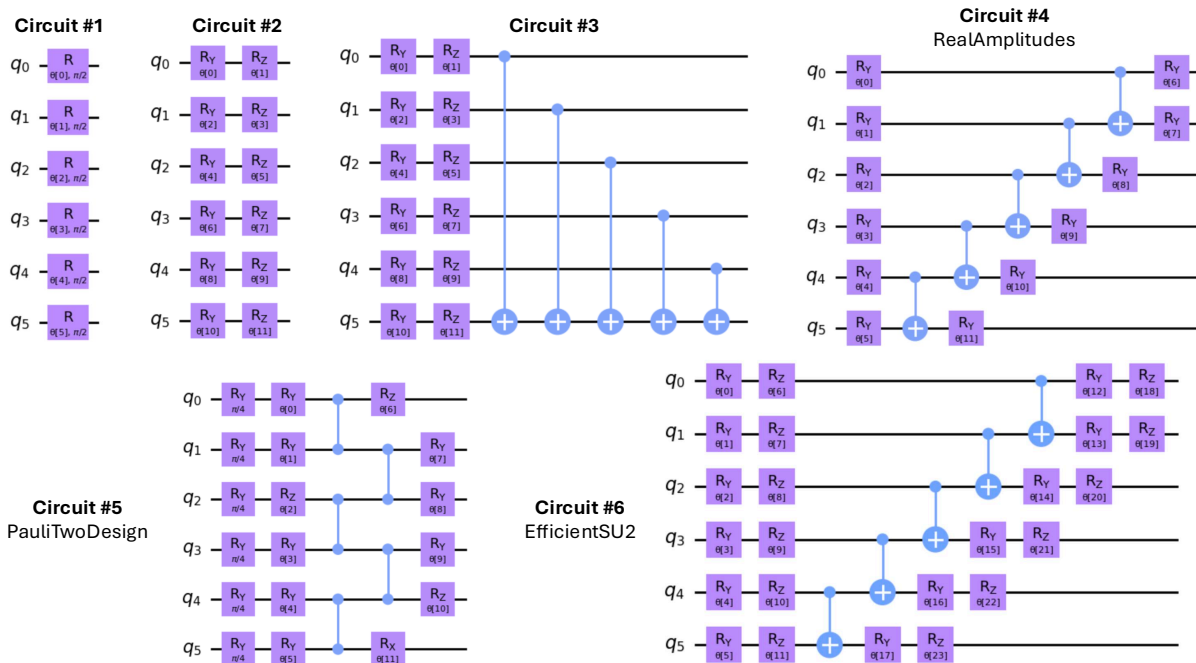


Figure 5: Visualization of Varied Circuits Rendered using Qiskit:³² Circuits obtained from JARVIS-tools³⁴ Circuits 4, 5, and 6 are also known as RealAmplitudes, PauliTwoDesign and EfficientSU2 circuits, respectively.

0.06 %) from the NumPy reference values across all three molecules. Also, we observe noticeably higher uncertainty values: Al^- has an uncertainty value of 0.061 264, 0.097 844 for Al_2^- , and 0.019 614 for Al_3^- (Table 2). This suggests that Circuit #5 might introduce some level of approximation or noise that affects its convergence accuracy. This observation makes sense because the PauliTwoDesign ansatz is generally known for its random entangling structure and for using a fixed design based on Pauli rotations.⁵³ While this circuit can be useful for exploring highly entangled states, its structure might introduce more variability or noise in energy convergence. Circuit #6 (EfficientSU2) on the other hand shows consistent results with zero deviation across all systems, highlighting the reasons why it was the default ansatz for ground-state energy calculations in the developed workflow. The slight variances between circuits indicate that the selection of the ansatz can influence the accuracy of VQE results. While most circuits perform similarly, the differences observed, especially in Circuit #5, emphasize the importance of carefully choosing a circuit structure that aligns with the

specific properties of the system being simulated.

Table 2: Comparison of Ground-State Energy (E_h) when Varying Circuit Type on Statevector Simulator, using Default Parameters: STO-3G Basis Set, SLSQP Optimizer, and 1 Repetition

(a) Al^-

	Al^- (VQE)	Variation (\pm)	Al^- (NumPy)
Circuit #1	-238.842 45	0.000 000	-238.870 47
Circuit #2	-238.842 42	0.000 000	-238.870 47
Circuit #3	-238.842 42	0.000 000	-238.870 47
Circuit #4	-238.842 39	0.000 000	-238.870 47
Circuit #5	-238.722 95	0.061 264	-238.870 47
Circuit #6	-238.842 31	0.000 000	-238.870 47

(b) Al_2

	Al_2 (VQE)	Variation (\pm)	Al_2 (NumPy)
Circuit #1	-477.799 05	0.000 000	-477.801 97
Circuit #2	-477.799 05	0.000 000	-477.801 97
Circuit #3	-477.799 04	0.000 000	-477.801 97
Circuit #4	-477.799 05	0.000 000	-477.801 97
Circuit #5	-477.529 26	0.097 844	-477.801 97
Circuit #6	-477.799 05	0.000 000	-477.801 97

(c) Al_3^-

	Al_3^- (VQE)	Variation (\pm)	Al_3^- (NumPy)
Circuit #1	-716.690 56	0.000 000	-716.708 73
Circuit #2	-716.690 54	0.000 000	-716.708 73
Circuit #3	-716.708 73	0.000 000	-716.708 73
Circuit #4	-716.708 73	0.000 000	-716.708 73
Circuit #5	-716.678 96	0.019 614	-716.708 73
Circuit #6	-716.675 25	0.000 000	-716.708 73

We also varied the number of reps, which refers to the number of times the parameterized ansatz layer is repeated within the quantum circuit. Increasing the number of reps allows the circuit to represent more complex states. We limited our analysis to only 1 to 4 reps, considering both system size and computational cost. We found that across all three systems (Al^- , Al_2 , and Al_3^-), the VQE yields energy estimates with less than 0.01 % difference than the reference NumPy values even with a minimal number of reps (1 rep) (Table 3). This

indicates that while increasing the number of repetitions can slightly improve accuracy, a simple ansatz structure can approximate the ground-state energy of these small systems well. Although additional reps (especially 3 and 4) slightly improve the energy values, the improvements are marginal. For example, in Al^- the VQE energy changes from -238.84231 at 1 rep to -238.84371 at 4 reps, a difference of about $0.0014 E_h$ (Table 3). Similarly, for Al_2 and Al_3^- the VQE energy values remain almost unchanged across reps, suggesting diminishing returns with additional repetitions. The uncertainty value remains either zero or negligible for all systems and reps, with very small non-zero values appearing only at higher reps. This stability implies that the solution does not fluctuate significantly with more repetitions, showing that the ansatz structure remains consistent across reps. As expected, job times increased significantly (14.1 seconds for 1 rep to 64.0 seconds for 4 reps for Al^-) with the number of reps (Table S1). Since the energy values are so close to the NumPy values with minimal reps, using a lower number of reps (1 or 2) would be more computationally efficient without sacrificing accuracy. Higher reps (3 or 4) provide only slight improvements, so they may be unnecessary for the studied systems.

We varied the simulator type, comparing results from the Statevector simulator and the QASM (Quantum Assembly Language) simulator, both of which are readily available on the Qiskit interface. The main difference between these two simulators is that the Statevector simulator computes the exact quantum state of a system and provides deterministic results without simulating measurement, making it ideal for theoretical analysis and debugging. The QASM simulator, on the other hand, simulates measurement outcomes by running circuits multiple times (shots), introducing shot-based statistical fluctuations. While it does not model hardware noise by default, its probabilistic outputs resemble the behavior of real quantum devices under repeated measurements. The Statevector simulator is faster for small systems, while the QASM Simulator provides a more realistic view of sampling-related variability in quantum computations.^{18,54}

In each case, the Statevector simulator produces VQE energy values closer to the refer-

Table 3: Comparison of Ground-State Energy (E_h) Results when Varying Number of Reps on Statevector Simulator, using Default Parameters: STO-3G Basis Set, SLSQP Optimizer, and Efficient SU2 Ansatz (Circuit #6)

(a) Al^-

	Al^- (VQE)	Variation (\pm)	Al^- (NumPy)
1 Rep	-238.842 31	0.000 000	-238.870 47
2 Reps	-238.842 48	0.000 000	-238.870 47
3 Reps	-238.857 88	0.000 001	-238.870 47
4 Reps	-238.843 71	0.000 007	-238.870 47

(b) Al_2

	Al_2 (VQE)	Variation (\pm)	Al_2 (NumPy)
1 Rep	-477.799 05	0.000 000	-477.801 97
2 Reps	-477.799 04	0.000 000	-477.801 97
3 Reps	-477.799 04	0.000 002	-477.801 97
4 Reps	-477.799 04	0.000 004	-477.801 97

(c) Al_3^-

	Al_3^- (VQE)	Variation (\pm)	Al_3^- (NumPy)
1 Rep	-716.675 25	0.000 000	-716.708 73
2 Reps	-716.708 72	0.000 000	-716.708 73
3 Reps	-716.708 73	0.000 000	-716.708 73
4 Reps	-716.708 72	0.000 000	-716.708 73

ence NumPy values compared to the QASM simulator (Table 4). This indicates that the Statevector simulator provides more accurate results, likely because it does not simulate shot-based statistical noise, unlike the QASM simulator. Results from the QASM simulator deviate more significantly from the NumPy reference values. This discrepancy is expected due to the probabilistic nature of the QASM simulator, which includes shot-based sampling, leading to slightly higher energy values for each molecule. As the size of the system increases (from Al⁻ to Al₃⁻), the difference between QASM and Statevector results appears to increase slightly. This trend could suggest that larger systems might experience compounded effects of noise in the QASM simulator, leading to greater deviations from exact values. Simulations on the QASM simulator also required significantly longer job times compared to the Statevector simulator, reflecting the additional computational burden of running multiple shots (Table S1). Despite these differences, the VQE calculations on both simulators yield energy results that remain within approximately 0.25 % of the NumPy reference values, indicating that the VQE algorithm, when optimized within the quantum-DFT embedding workflow, can produce stable energy estimates even in the presence of shot-based statistical noise. These results emphasize the differences between using an ideal, noise-free simulation (Statevector) versus a more realistic but noisier simulation (QASM). It also highlights the ability of the VQE approach in estimating ground-state energies under different computational conditions.

Table 4: Comparison of Ground-State Energy (E_h) Results of all Studied Systems with Varied Quantum Simulator Type (Statevector vs. QASM Simulator). Default Parameters including a STO-3G Basis Set, the SLSQP Optimizer, an Efficient SU2 Ansatz (Circuit #6), and 1 Repetition

	Al ⁻ (VQE)	Al ⁻ (NumPy)	Al ₂ (VQE)	Al ₂ (NumPy)	Al ₃ ⁻ (VQE)	Al ₃ ⁻ (NumPy)
Statevector	-238.842 31	-238.870 47	-477.799 05	-477.801 97	-716.675 25	-716.708 73
QASM	-238.310 78	-238.870 47	-477.206 89	-477.801 97	-716.470 32	-716.708 73

We evaluated the performance of several basis sets—Slater-Type Orbital with 3 Gaussians (STO-3G), 3 Gaussian primitives with 2-1 split valence (3-21G), Slater-Type Orbital with 6 Gaussians (STO-6G), 6 Gaussian primitives with 3-1 split valence (6-31G), correlation-consistent polarized valence double-zeta (cc-pVDZ), and correlation-consistent polarized va-

lence triple-zeta (cc-pVTZ), ordered from lowest to highest level complexity — for estimating ground-state energies of aluminum systems Al^- , Al_2 , and Al_3^- using the VQE algorithm (Tables 5). As expected, job times increased (from 14.1 seconds using STO-3G to 60.0 seconds using cc-pVTZ for Al^-) as the basis sets became more complex (Table S1). We note that the energy values calculated with the larger basis sets (cc-pVTZ for Al_2 and cc-pVDZ/cc-pVTZ for Al_3^-) were not obtained due to lack of memory. We also utilized the obtained energy values of Al_2 for comparison against published results on the CCCBDB,⁵⁵ which were calculated using DFT and LDA (Table 6). This comparative data is important to this study because it demonstrates that the developed quantum-DFT embedding workflow yield results that align with classical references, supporting the utility of VQE in specific chemistry and materials applications.

The results show a general trend across the basis sets: as the basis set quality increases, the VQE energy values approach the NumPy reference values, indicating improved accuracy with more complex basis sets. For instance, in the Al^- system, STO-3G provides a VQE energy of $-238.842\ 31\ E_h$, while cc-pVDZ gives a closer approximation at $-241.860\ 32\ E_h$, aligning with the NumPy reference of $-241.866\ 67\ E_h$ (Table 5). Similarly, for Al_2 and Al_3^- , the higher-level basis sets (6-31G and cc-pVDZ) yield results that are closer to the reference values, underscoring the importance of basis set choice in capturing accurate ground-state energies for aluminum clusters. When analyzing the percent errors among the basis sets in comparison to CCCBDB (Table 6), we see a consistent decrease in error as the basis set quality improves. For Al_2 , the STO-3G basis set produces a percent error of 0.1369 %, whereas the cc-pVDZ basis set reduces this error to 0.1119 %. This trend suggests that while minimal basis sets like STO-3G are computationally efficient, they sacrifice some accuracy. In contrast, the larger and more complete basis sets, such as cc-pVDZ, offer more reliable estimates of electronic energies, providing an optimal balance between accuracy and computational demand for these VQE calculations. Table 6 adds an important layer of analysis by comparing our VQE and NumPy results to published data from the CCCBDB.

For each system and basis set, the CCCBDB provides benchmark energy values against which we can evaluate the accuracy of our calculations. As we move to higher-level basis sets, such as 6-31G and cc-pVDZ, both the VQE and NumPy results align more closely with the CCCBDB values, with errors consistently reducing. This validation against CCCBDB highlights that our VQE methodology, especially with the cc-pVDZ basis set, achieves ground state energy estimates comparable to published data.

Table 5: Comparison of Ground-State Energy (E_h) Results when Varying Basis Sets on Statevector Simulator, using Default Parameters: SLSQP Optimizer, Efficient SU2 Ansatz (Circuit #6), and 1 Repetition

(a) Al⁻

	Al ⁻ (VQE)	Variation (\pm)	Al ⁻ (NumPy)	Variation (\pm)
STO-3G	-238.842 31	0.000 000	-238.870 47	0.000 000
3-21G	-240.540 13	0.000 406	-240.547 58	0.000 655
STO-6G	-240.800 47	0.000 020	-240.826 96	0.000 479
6-31G	-241.844 63	0.000 803	-241.851 93	0.000 480
cc-pVDZ	-241.860 43	0.000 235	-241.867 21	0.000 908
cc-pVTZ	-241.860 32	0.000 039	-241.866 67	0.000 072

(b) Al₂

	Al ₂ (VQE)	Variation (\pm)	Al ₂ (NumPy)	Variation (\pm)
STO-3G	-477.799 05	0.000 000	-477.801 97	0.000 000
3-21G	-481.076 34	0.012 279	-481.095 08	0.013 676
STO-6G	-481.710 46	0.000 000	-481.713 39	0.000 000
6-31G	-483.703 54	0.000 732	-483.706 66	0.010 760
cc-pVDZ	-483.737 01	0.011 890	-483.741 61	0.010 836

(c) Al₃⁻

	Al ₃ ⁻ (VQE)	Variation (\pm)	Al ₃ ⁻ (NumPy)	Variation (\pm)
STO-3G	-716.675 25	0.000 000	-716.708 73	0.000 000
3-21G	-721.694 50	0.000 000	-721.664 45	0.377 995
STO-6G	-722.540 74	0.000 000	-722.575 00	0.000 000
6-31G	-725.601 86	0.000 000	-725.603 34	0.000 000

The comparison to CCCBDB data confirms that our VQE-based approach can approximate ground-state energies with errors within 0.2 % for small aluminum clusters under simulation (Table 6). While minimal basis sets like STO-3G can provide rough estimates,

Table 6: Ground-State Energy Results of Al_2 (E_h) on Statevector Simulator with Varied Basis Sets, using Default Parameters: SLSQP Optimizer, Efficient SU2 Ansatz (Circuit #6), and 1 Repetition in Comparison to CCCBDB Values

Basis Set	VQE Energy	Variation (\pm)	Percent Error	NumPy Energy	Variation (\pm)	Percent Error	CCCBDB
STO-3G	-477.799 05	0.000 000	0.1369 %	-477.801 97	0.000 000	0.1375 %	-477.145 74
3-21G	-481.076 34	0.012 279	0.1153 %	-481.095 08	0.013 676	0.1192 %	-480.522 53
6-31G	-483.703 54	0.000 732	0.1133 %	-483.706 66	0.010 760	0.1139 %	-483.156 31
cc-pVDZ	-483.737 01	0.011 890	0.1119 %	-483.741 61	0.010 836	0.1129 %	-483.196 25

using higher-level basis sets such as 6-31G and cc-pVDZ significantly improves the accuracy of our results, bringing them closer to established benchmark values. This analysis underscores the importance of basis set selection in quantum-DFT embedding workflows, especially for applications requiring high precision, as the choice of basis set directly impacts the reliability of energy estimations.

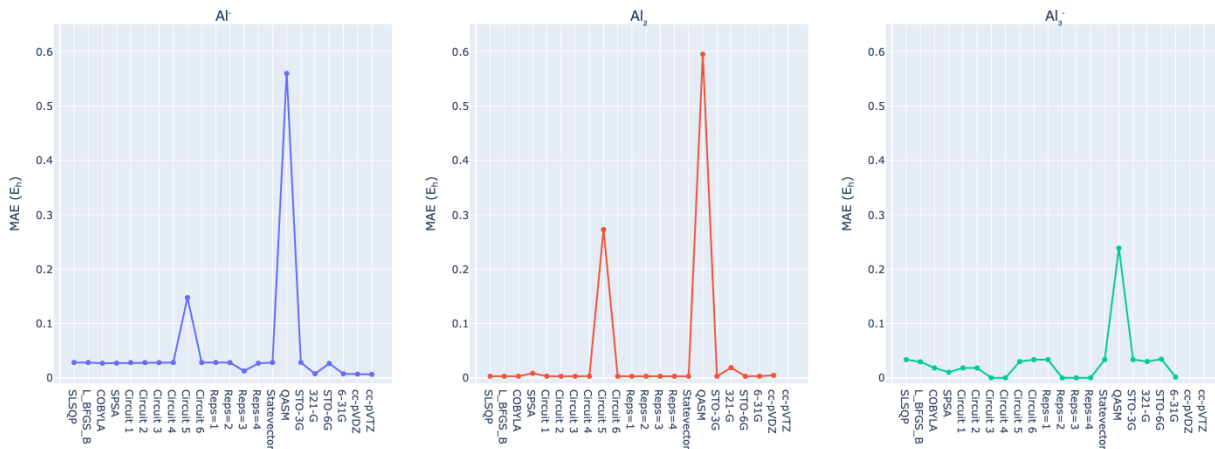


Figure 6: Mean Absolute Energy (in Hartree) of Calculated VQE Values in Comparison to NumPy Values of Al^- , Al_2 , and Al_3^- . This figure summarizes results across six parameter types: classical optimizer, ansatz circuit, ansatz depth (reps), simulator type, and basis set. The sharp error spikes correspond to known outliers (e.g., PauliTwoDesign, QASM).

The standard benchmark for chemical accuracy is approximately 1 kcal/mol, or 0.0016 Hartree. While some of the absolute energy errors in our VQE simulations exceed this threshold, the relative percent errors remain below 0.2 % in comparison to CCCBDB. These results are promising given the use of a low-depth, hardware-efficient ansatz and NISQ-compatible constraints. The observed trends, especially with higher-level basis sets like cc-

pVDZ, suggest that further improvements in ansatz design, noise mitigation, and embedding strategy could bring future simulations closer to this standard.

To summarize the benchmarking results across all studied parameters, we present in Figure 6 the mean absolute error (MAE) between VQE energy estimates and corresponding NumPy reference values for each configuration across Al^- , Al_2 , and Al_3^- . This consolidated view allows for direct comparison of how classical optimizers, circuit types, ansatz repetitions, simulator types, and basis sets influence VQE accuracy. Notably, the largest deviations were observed for the PauliTwoDesign circuit (Circuit #5) and the QASM simulator, consistent with earlier tabled results. These outliers significantly exceed the commonly accepted threshold for chemical accuracy. In contrast, most other configurations yield MAEs close to or below this threshold, reinforcing the utility of certain low-depth circuits and optimizer choices under simulated conditions. This figure complements the detailed numerical tables provided in the Supporting Information, offering an intuitive visual overview of VQE performance and helping to identify configurations that deliver both accuracy and computational efficiency.

Lastly, to further evaluate the performance of the developed workflow under realistic conditions, we compared the VQE results for Al_2 obtained using noise models to the CC-CBDB benchmark values (Table 7). Due to limited access to actual quantum hardware, we employed five IBM simulated noise models—FakeSherbrooke, FakeManhattan, FakeToronto, FakeTokyo, and FakeMontreal—to approximate the effects of noise, such as gate errors, decoherence, and measurement inaccuracies. These models were integrated into the QASM simulator, which inherently introduces shot-based statistical fluctuations through repeated measurements. When combined with noise models, the QASM simulator enables realistic simulations that capture both sampling uncertainty and device-level noise effects. According to IBM Quantum Documentation, noise models mimic the behaviors of quantum hardware by using snapshots, which contain critical information such as the coupling map, basis gates, and qubit properties. These details are crucial for performing noisy simulations that closely

resemble real quantum hardware.³² While these models do not fully capture real-device performance, they serve as an approximation of hardware-level noise under QASM simulation - making them an essential tool for assessing quantum algorithms and workflows in the NISQ era.

Table 7: Ground-State Energy Results of Al₂ (E_h) on the QASM Simulator including Noise Models, using Default Parameters: STO-3G Basis Set, SLSQP Optimizer, Efficient SU2 Ansatz (Circuit #6), and 1 Repetition in Comparison to CCCBDB Values

Noise Model	VQE Energy	CCCBDB Energy	Percent Error
FakeSherbrooke	-477.211 77	-477.145 74	0.01384 %
FakeManhattan	-477.173 84	-477.145 74	0.00589 %
FakeToronto	-477.220 75	-477.145 74	0.01572 %
FakeTokyo	-477.192 91	-477.145 74	0.00989 %
FakeMontreal	-477.203 17	-477.145 74	0.01204 %

The VQE energy estimates show minimal deviation from the CCCBDB reference energy of $-477.145\ 74\ E_h$, with percent errors ranging from 0.005 89 % (FakeManhattan) to 0.015 72 % (FakeToronto). This consistency across noise models highlights the robustness of the VQE algorithm and the effectiveness of the quantum-DFT embedding workflow in producing reliable results under noisy conditions. Among the noise models, FakeManhattan exhibited the smallest deviation, suggesting that it most accurately replicates the performance of real hardware for this particular system. Conversely, FakeToronto showed the largest percent error, likely due to variations in its simulated error rates. Despite these differences, all results remained less than 0.02 %, emphasizing the capability of the workflow to handle noise while delivering accurate energy estimates. This comparison highlights the practical utility of noise models for evaluating algorithm behavior under hardware-like conditions, especially when access to quantum devices is constrained. The agreement of the noise model results in comparison to the CCCBDB benchmark highlights the effectiveness of the VQE algorithm within the developed workflow and reinforces its potential utility in evaluating quantum chemistry benchmarks under simulated conditions. These findings are especially promising for future applications, as they suggest that similar accuracy can be achieved on

real quantum devices as hardware continues to improve.

Incorporating noise models into benchmarking studies when access to hardware is limited is essential for evaluating quantum algorithms under conditions that reflect near-term hardware constraints. As quantum hardware remains constrained by noise and limited qubit counts, noise-aware benchmarking provides critical insights into algorithm performance and helps identify areas for improvement. Future work could extend this analysis to other molecular systems in the CCCBDB to further validate the applicability of the workflow. Also, studying error mitigation techniques within noise models could help further narrow the gap between simulated and experimental results, enhancing the reliability and applicability of quantum computations in materials discovery.

Conclusions and Future Perspectives

This study demonstrates the applicability of the Variational Quantum Eigensolver (VQE) within a quantum-DFT embedding framework (on a quantum simulator) for simulating the ground-state energies of aluminum clusters, with applications to materials discovery and design. By systematically benchmarking key parameters, including classical optimizers, circuit types, number of repetitions, simulator types, and basis sets, we achieved insights into optimizing VQE’s performance on quantum simulators with and without added noise. Notably, our findings show that SLSQP and COBYLA optimizers yield efficient convergence, while EfficientSU2 circuits and minimal repetitions provide reliable results without excessive computational cost. Furthermore, basis set selection proved critical; higher-level basis sets like cc-pVDZ enhanced accuracy, aligning our VQE results with NumPy benchmarks and published data from the CCCBDB.

Although real-device execution was not included due to resource constraints, the use of noise models allowed for a preliminary assessment of robustness under NISQ-relevant conditions. The results showed minimal deviation from the CCCBDB benchmark, with percent

errors consistently below 0.02 % across the five IBM noise models tested. The noise model results suggest the developed workflow is robust in simulation and may yield comparable performance on real devices as hardware capabilities improve. Among the noise models, FakeManhattan demonstrated the smallest error. Our analysis indicates that VQE can produce comparable results in the presence of simulated noise for small systems. While these results are promising, they are obtained from simulations under controlled conditions. Additional benchmarking on real quantum devices and comparisons with alternative embedding approaches remain necessary to establish broader generalization.

Looking forward, our work lays the foundation for expanded benchmarking and tool development to support quantum chemistry and materials science research. Future efforts will involve testing the workflow on real devices and exploring error mitigation strategies. We also plan to benchmark our DFT embedding approach against other quantum embedding techniques such as DMET, to evaluate their respective trade-offs in accuracy and resource efficiency. Expanding the benchmarking to a broader range of chemical systems beyond aluminum clusters will support the generalizability of our findings. Additionally, we aim to integrate our findings into the JARVIS-AtomQC platform, making results readily accessible to the broader research community. Finally, we plan to develop a dedicated Python package that encapsulates our benchmarking workflow, enabling researchers to efficiently test and optimize quantum algorithms for material simulations. These advancements will contribute to establishing standardized practices in quantum algorithm benchmarking, accelerating progress toward practical quantum computing applications in materials discovery and beyond.

Data Availability

The code and data used in this work are available at <https://github.com/usnistgov/BenchQC>.

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Supporting Information Available

Please see Supporting Information for additional data tables including specific energy values for all systems.

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TOC Graphic

