

# Deloitte Quantum Challenge 2023

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## Abstract

In this project we explored VQE and ADAPT-VQE to simulate  $CO_2$ ,  $N_2$  and  $H_2O$  capture by an Cu(I) ion. We also simulated the capture of  $CO_2$  in the presence of  $N_2$ . We explored ansatz selection and how different ansatz give rise to different results. Our results are in line to the ones presented in the Quantistry Platform. We also present an escalation of these algorithms, that can be sensibly run with 25800 physical qubits, a value that should be achieved in the coming years.

## 1 Team

The team - The Entangled Cats - is composed by a single member: José Diogo da Costa Jesus. José is a Portuguese physicist, having recently completed a masters in Engineering Physics from Instituto Superior Técnico, University of Lisbon. His main focus currently is quantum computing and information, having already participated in numerous competitions and hackathons. For contact please refer to the following email: josdcjesus@gmail.com.

## 2 Challenge definition

The development of Direct Air Capture (DAC) of Carbon is seen by many as a strategic technology to fight Climate Change. Indeed, this development would allow us to remove  $CO_2$  directly from the air and correct not only legacy emissions, but also provide a means to reduce emissions from sources where green transitions are underdeveloped or are not feasible.

One of the crucial ingredients in DAC is the development of appropriate filters for carbon capture. Indeed, DAC methods are very energy intensive, due to the low concentration of  $CO_2$  in the atmosphere, which implies the need for large fan operation, as well as the need for filter regeneration, which requires cycles of heating the filters to release the  $CO_2$  so that it can be stored. Therefore, the development of new filters, capable of large absorption of  $CO_2$  is seen as crucial.

One type of molecular structure with a lot of potential as a DAC filter, are the Metal-Organic Frameworks (MOFs). They are generally crystalline compounds constituted by metal ions, or clusters of metal ions, and organic and organic ligands forming 3 dimensional porous structures. In particular, they possess high energy surface area, tunnable pore size and low heat capacity, all desirable properties in a DAC filter.

This way, in order to design cost effective MOF for DAC, electronic and structural information obtained via computational chemistry is crucial. However, it is well known that current computational methods are based on approximations that quickly become flawed with increasing complexity. This, is where Quantum Computers and Algorithms may have a distinct advantage, for they use Nature's quantum properties to simulate quantum effects.

This way, this years Deloitte's Quantum Climate Challenge focus on exploring the feasibility of accelerating carbon capture MOF development using Quantum Computers. This challenge is divided into 2 tasks: Task 1 focus on efficiently calculating the potential energy surface of a system of MOF constituents and atmospheric gas molecules, exploring which calculations are already possible on today's quantum computers and comparing them to classical quantum chemistry calculations and Task 2 focus on developing a scheme to calculate the carbon dioxide uptake and evaluating the potentially accelerated development of MOFs for DAC when using more advanced quantum computers in the future.

## 3 Task 1

### 3.1 Task 1 A

The objective in task 1 A is to create a quantum algorithm, a quantum hybrid or quantum inspired solution, that calculates the minimum of the Potential Energy Surface (PES) of combinations gas molecules and ions. To that objective, the participants must choose at least an ion and obtain the PES for at least Carbon dioxide and one more molecule.

#### 3.1.1 VQE and UCCSD Ansatz

As a starting approach to this task we followed the guidelines and used the Variational Quantum Eigensolver (VQE) to obtain the PES of Carbon Dioxide Capture by Cu(I). This ion was chosen because the Quantistry platform provided VQE references for its reaction with  $CO_2$ ,  $H_2O$  and  $N_2$ .

The Variational Quantum Eigensolver is an hybrid algorithm that leverages classical optimization to improve an initial Ansatz, or guess, of the ground state of the system. This ground state is prepared in a Quantum Computer that also measures its energy relative to the problem Hamiltonian.

In a first approach, we followed the guidelines and selected the UCCSD Ansatz. This Ansatz is a staple of quantum chemistry and simulates all possible single and double electron excitations in the system (therefore not changing the electron number just as expected in a real chemical reaction). To this we coupled as an initial starting point the Hartree Fock state.

Increasing the complexity relative to the guidelines, instead of using "STO-3G" as our basis set we used the "cc-pVDZ" set, as was used in the Quantistry calculations (in fact using the "STO-3G" set always produced an error of set of a few thousand eV relative to the Quantistry reference). Additionally, instead of using the Simultaneous Perturbation Stochastic Approximation (SPSA) optimizer, we used the Constrained Optimization By Linear Approximation (COBYLA) optimizer for this shower to be faster and to take less iterations to converge to a result.

Finally, instead of using following the Guidelines and using the Jordan-Wigner mapping between fermionic operators to qubit operators we used the Parity mapper, readily available on Qiskit [1]. The main motive for using this mapper is that it allows us to use 2 qubit reduction [2] and therefore remove 2 qubits from our simulations.

Additionally, we also had to consider some specific active space for our simulations. Indeed, taking a direct approach and simulating all electrons in the system would be infeasible at the moment, for it would require 66 qubits (66 spin orbitals). Therefore, it is much more interesting to consider just an active space with few active electrons that still capture the dynamics of the system, whilst using classical approximations for the rest. This approach was also the one used in the Quantistry references, where it was considered an UCCSD ansatz with an active space of 6 electrons and 6 active orbitals (UCCSD(6,6)).

This way, as a starting point for the challenge, we also considered an UCCSD(6,6) ansatz. However the results were not encouraging. Using an ideal simulator with 10000 shots per circuit the results obtained were:

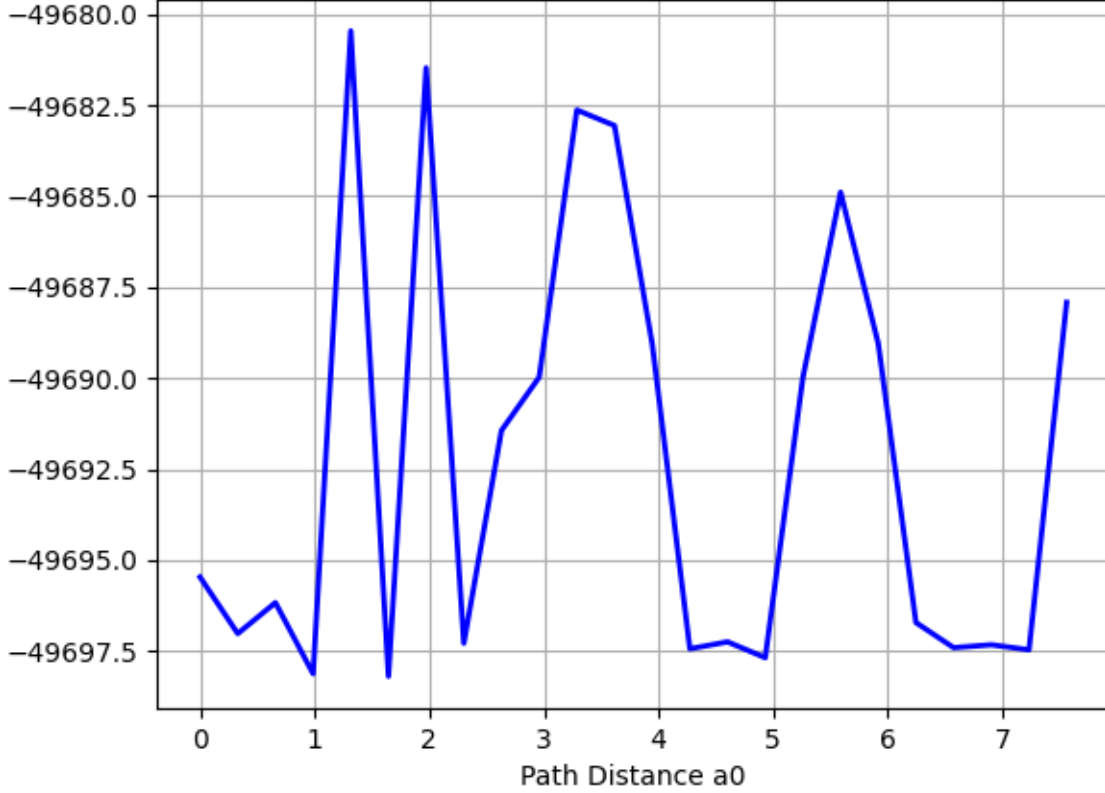


Figure 1: Computed Energy (eV) vs Path distance for the UCCSD(6,6) ansatz

From these calculations, it became clear that the UCCSD ansatz is far too complex to be run with this active space. This is even more obvious if one takes into consideration the effects of noise. Indeed, this ansatz uses 3700 CNOT gates, which being 2 qubit gates are usually the main source of error, a number that is far too large to be run effectively in today’s quantum computers. Therefore a better approach was needed.

### 3.1.2 ADAPT-VQE

Instead of simply using VQE, as suggested in the guidelines, we took a more complex approach and instead explore the more advanced ADAPT-VQE algorithm [3].

ADAPT-VQE is a quantum/classical algorithm with the same objective as VQE but with the added advantage of automatically selecting and creating a high performance Ansatz. Indeed, in this algorithm we first select a set of possible gates and a desired accuracy. Iteratively, the algorithm will select the gradients that produce the largest gradients in our cost function. This way, we can create a much shallower Ansatz that can still be efficiently optimized and reach the ground state.

This algorithm was implemented using Qiskit resources. We first applied it to calculate the PES for carbon dioxide capture by  $Cu(I)$ . The results obtained were:

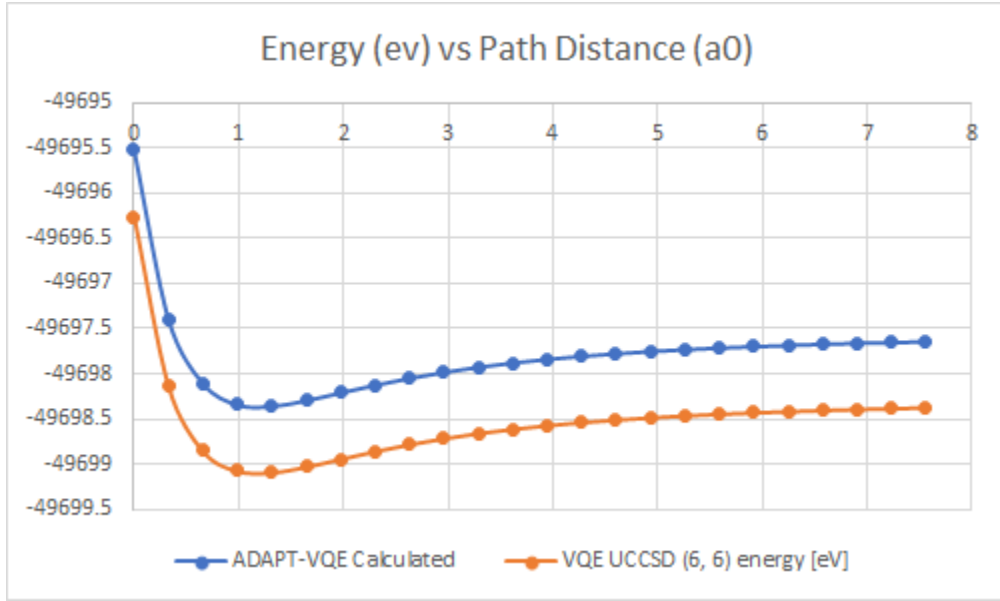


Figure 2: Computed Energy (eV) vs Path distance for the Ansatz computed for the (4,4) active space for  $CO_2$  capture by  $Cu(I)$

where we used UCCSD(4,4) as our starting point. As we can see the obtained results are in line with the reference provided in the Quantistry platform.

The smaller subspace has the advantage of needing less qubits and CNOT gates which implies a speedup on the algorithm application and less noise in actual applications on Quantum Computers.

Using the smaller subspace we also applied the same Ansatz to other gas molecules. While at first one would think that a new optimization process would be required, we verified that the Ansatz actually performs quite well for calculating the PES of capturing  $N_2$  and  $H_2O$ . This intuitively means that our Ansatz will be most dependent on the metal ion chosen. The results obtained were:

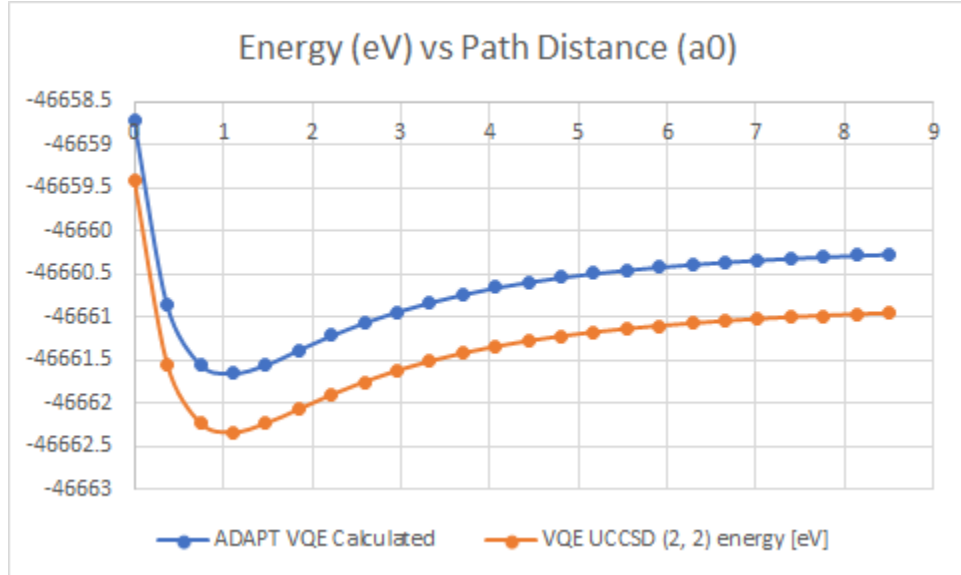


Figure 3: Computed Energy (eV) vs Path distance for the Ansatz computed for the (4,4) active space for  $H_2O$  capture by  $Cu(I)$

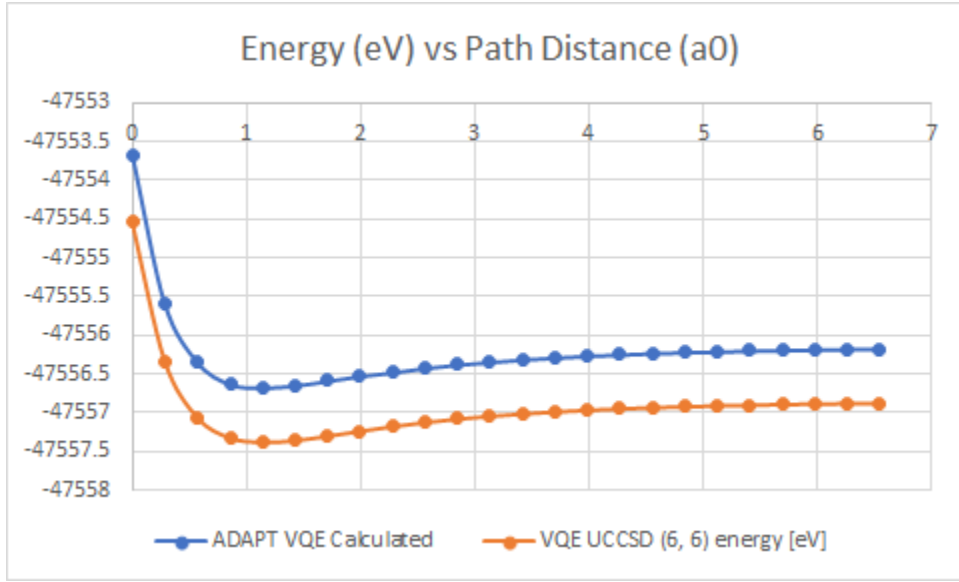


Figure 4: Computed Energy (eV) vs Path distance for the Ansatz computed for the (4,4) active space for  $N_2$  capture by  $Cu(I)$

Finally, to increase complexity we also considered for  $Cu(I)$  a case where one extra gas molecule of  $N_2$  is at an ideal distance from the metallic ion and a  $CO_2$  molecule is placed at a specific distance from the ion and the gas molecule. We studied the PES in terms of distance from  $CO_2$  to the metallic ion and the angle between the gas molecules. The results were:

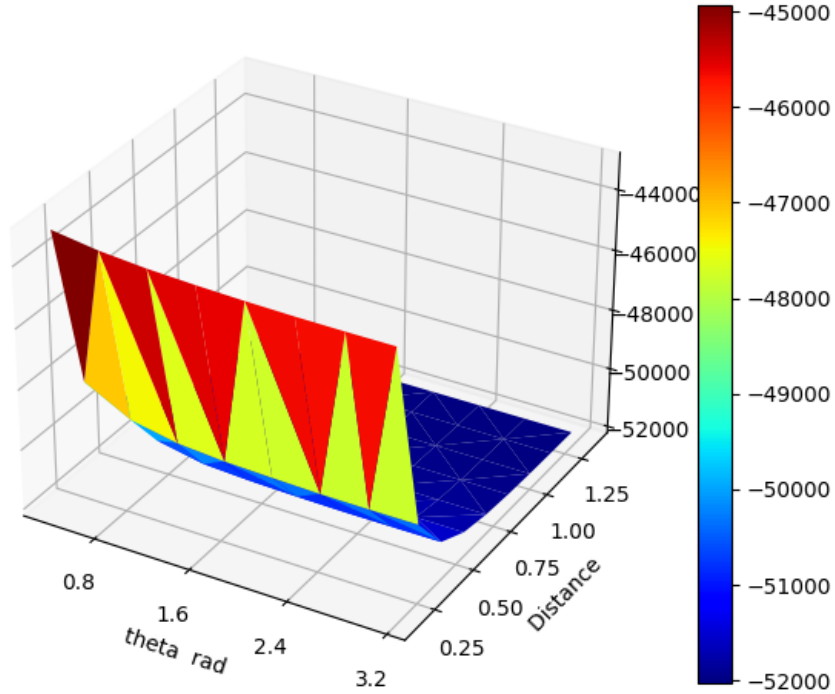


Figure 5: Computed Energy (eV) vs Path distance (Å) between  $Cu(I)$  and  $CO_2$  and angle between  $CO_2$  and  $N_2$  (rad)

This results suggests that unfortunately if the ion is already bound to  $N_2$ , the  $CO_2$  will not be captured. However, this simulation is quite shallow ( for instance it does not take into consideration

parts of the organic linker) and therefore should not be used as an argument against  $CO_2$  filters made from Cu(I).

### 3.2 Task 1 B

We can now compare our solution to the classical solutions presented in the Quantistry Platform. First and foremost, the biggest advantage of our solution is, although we are currently still relying on classical approximations, given more powerful quantum computers, we could potentially remove all these approximations and have a true quantum simulation, something that is completely out of reach when considering classical approaches.

Secondly, it must be noted that by only considering the parts of the Ansatz that produce the greatest gradients we can actively speed up the method a great deal. Indeed, for the  $CO_2$  capture simulation, we managed to run the whole thing in about 3 minutes, a considerable speed up to the 13.51 core hours presented in the Quantistry Platform. The  $N_2$  and  $H_2O$  took around the same time.

However there are 2 main disadvantages regarding our simulations. The first is that, because we considered a reduced Active Space instead of the entirety of the available orbitals we effectively neglect quite a lot of electron-electron effects (which reduces the post Hartree-Fock components of our solutions). Although this is still an improvement when compared to Hartree Fock methods, we can see that our solution lacks when compared to more advanced methods such as MP2 Energy or CCSD Energy.

Finally, the last drawback is related to errors in calculations. Indeed current quantum computers are still quite noisy and when we take in consideration this noise our solutions worsen quite dramatically, even when using advanced noise mitigation methods such as Zero Noise Extrapolation (ZNE [4]).

Nonetheless, for small systems our solution produces faster and more accurate simulations than traditional Hartree Fock calculations which demonstrates the potential that Quantum Computers will bring to chemistry in the future.

## 4 Task 2

### 4.1 Task 2 A

In this task, the challenge was to conceptualize an algorithm to scale the calculation from one binding site to at least one 2D unit cell of the given metal organic framework-family AND from one gas molecule to a larger amount of substance of the gas molecule. This way, taking into consideration the results from the previous task we considered the following approach:

- Step 1 - Considering 1 unit 2D cell, place a  $CO_2$  molecule in each;
- Step 2 - obtain the corresponding UCCSD Ansatz with at least 4 active electrons and 4 active orbitals per binding site (so using 2 qubit reduction that gives 36 qubits);
- Step 3 - compute the gradients for each active operation in the Ansatz for their optimal points;
- Step 4 - select the 12 operations that possessed the largest gradient (in line with task 1 - 2 operations per binding site) and Build a new Ansatz
- Step 5 - Run VQE algorithm with this ansatz dependent on the path distance for all gas molecules to their respective binding site

If the minimum computed energy is not negative, i.e, if the reaction is not thermodynamically viable, we go back to step 1 and remove 1 gas molecule and so far until we find the maximum amount of  $CO_2$  that can be caught. Alternatively, one can also consider different gas species and see how they interact with the cell and each other.

The main limitation of this solution is that we only take the operations that generate the greatest gradients. Obviously this can be changed by considering more operations, however a delicate balance need to be taken into consideration between additional error from noise and a bigger, more complete ansatz. The same logic applies to the number of qubits, if we do not restrict ourselves to active spaces and look at all available orbitals we will be much more susceptible to noise.

Finally, one last limitation to take into account is that this proposal does not take into consideration effects between cells, that although weak, could have a real impact in the filter's performance.

## 4.2 Task 2 B

In this task the challenge was to consider the requirements for an implementation of the concept from task 2A in real Quantum Computers. Well, it will all depend on the size of the active space and how resistant to noise we want our solution to be.

Indeed, if we consider only 4 active electrons and 4 active orbitals per binding size we would need 36 electrons. This can actually be implemented in a Quantum Computer today! However this does not take into consideration error that although mitigated, can not be corrected.

In order to implement error correction we need logical qubits. If we take the minimum number of qubits required to implement fault-tolerant logical qubits, which is 5, we would need a total of 180 qubits. Again this can also be computed in today's quantum computers. An example of that is IBM's Osprey processor with 433 qubits.

However, if we want to take into consideration the entire 2D cell, a lot more qubits are required. Indeed, considering the 2D cell built of Cu(I) presented in the Quantistry platform, with all binding sites active would require a simulation of 3528 orbitals - which requires 7056 logical qubits and 35280 physical qubits.

This however is quite overkill, and it is much more sensible to use a freeze core approximation and take into consideration only the valence orbitals. That gives us a total of 2580 orbitals - which requires 5160 logical qubits and 25800 physical qubits.

These values are quite high. However we are not taking into consideration techniques to reduce the size of the system, such as entanglement forging, that allows us to run 2n qubit algorithms with n qubits Quantum Computers. Nevertheless, steady progress is being made and a 26000 qubit quantum computer might be on the horizon given IBM's road-map that predicts 10s of thousands of qubits as soon as 2026 [5].

## 5 Conclusion

In this project we employed techniques to simulate Carbon capture, namely VQE and ADAPT-VQE. This allowed us to improve our understanding of quantum chemistry and the importance of ansatz selection when running variational algorithms. Our results in task 1A are very much in line with the expected from the Quantistry Platform.

We also proposed an escalation of our algorithm to an entire 2D cell. This proposal has advantages and disadvantages, but given its requirements should be possible to run in the next few years.

To conclude, this challenge was a wonderful learning opportunity and allowed the team to get a feel for the real utility of Quantum Computers.

## References

- [1] Qiskit. <https://qiskit.org/>.
- [2] 2 qubit reduction. <https://qiskit.org/documentation/stubs/qiskit.opflow.converters.TwoQubitReduction.html>.
- [3] Adapt-vqe. <https://qiskit.org/documentation/stable/0.25/stubs/qiskit.chemistry.algorithms.AdaptVQE.html>.
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- [5] Ibm quantum roadmap. <https://www.ibm.com/quantum/roadmap>.