

# Computational insights into CO<sub>2</sub> binding energy with a new iron(II) vinylic polyamine complex for exploratory CO<sub>2</sub> reduction studies

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## Abstract

Catalytic carbon dioxide (CO<sub>2</sub>) reduction proposes solutions to many environmental concerns. So far, iron-quaterpyridine complexes are among the forefront of the best candidates for CO<sub>2</sub>-to-CO conversion. Herein, a vinylic polyamine, (1Z,1'Z)-N1,N1'-((Z)-ethene-1,2-diyl)bis(ethene-1,2-diamine) (ZE-DiAm) is proposed as a new ligand alternative to quaterpyridine, which offers less structural complexity and comparable ligand position. Notably, ZE-DiAm also possesses higher basicity, which leads to a more stable complex. The study was carried out using density functional theory (DFT) computation to calculate the theoretical binding energy of CO<sub>2</sub> to [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup> as the representation of iron-ZE-DiAm complex. The binding energy was found to be 262.262 kJ/mol which is higher than typical binding energies calculated for Fe(II)-CO<sub>2</sub> interaction that ranging between 33.890 kJ/mol and 125.520 kJ/mol. This result implies that the complex [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup> forms a more stable bond with CO<sub>2</sub>, thus increasing its potential as a catalyst in the activation of CO<sub>2</sub>. This investigation, therefore, sets the basis for the further laboratory works on catalytic properties of [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup> in order to promote reductive transformations of CO<sub>2</sub> to CO and other reduced products.

**Keywords:** CO<sub>2</sub> conversion, iron complexes, ZE-DiAm, DFT Analysis, binding energy

## 1. Introduction

The prominence role of CO<sub>2</sub> in global warming and climate change results from it being the most concentrated greenhouse gas in the atmosphere. It has played an active part in the greenhouse effect for the past 200 years [1]. Burning of fossil fuels plus deforestation are a couple of the human activities that have greatly increased atmospheric CO<sub>2</sub> levels ever since the industrial revolution. As of 2023, atmospheric CO<sub>2</sub> levels have exceeded a concerning concentration – 420 parts per million (ppm). [2]. This enormous increase from the pre-industrial revolution levels of approximately 280 ppm [3] is concerning. CO<sub>2</sub>'s heat trapping ability in the atmosphere is reflected in higher global average temperatures, decreasing ice area in both poles, and more common extreme weather events such as hurricanes and storms. Reducing

atmospheric CO<sub>2</sub> levels will help to mitigate and stabilize the climate. Switching over to renewable energy sources, beefing up carbon capture technology, and preserving forests are some of the efforts needed to bring a slower pace of global warming as well as to ensure a future planet for us.

Strategies in cutting down carbon emissions are now largely focused on venting, storing, and converting these excess CO<sub>2</sub>. Among those strategies, carbon capture and storage (CCS) is one of mature technologies that has been widely used in industries. The captured CO<sub>2</sub> is either stored underground or mineralized which provides a way to prevent the re-entry of CO<sub>2</sub> into the atmosphere [4]. Apart from CCS, carbon conversion technologies are on a rising trend, and there is an increasing importance as efforts to turn the captured CO<sub>2</sub> into reduced products that are more valuable such as carbon monoxide (CO), methanol, hydrocarbons, or even elemental carbon. Electrochemical CO<sub>2</sub> reduction is as well getting many attentions because it opens the possibility to turn CO<sub>2</sub> into valuable products using renewable electricity [5]. Catalysis is central to these conversions, especially in the case of reducing CO<sub>2</sub> to CO which is an important starting material for various industrial applications, including synthetic fuels and chemical production. The value of converting CO<sub>2</sub> to CO is that it can be directly connected to existing chemical processes, allowing the carbon emissions to be recycled by industries and reducing overall environmental impact [6]. Developing efficient catalysts is still very important for making these technologies energy-efficient and scalable [7].

The exploration of new ligands is crucial for the advancement of CO<sub>2</sub> reduction technology to design systems which are both efficient and practical. To find ligands that have a lot of structural simplicity yet exhibit very good catalytic performance is plausible. Effective ligands must be able to show strong stability under reaction conditions, appropriate basic strength aiding activation of CO<sub>2</sub>, working together with metal centers. Also, they must be coordinating-versatile to ensure they effectively coordinated to metal atoms. In CO<sub>2</sub> reduction processes, ligands often play a crucial role to stabilize transition-states and have a large effect on the electronic environments surrounding metal catalysts which in turn affects efficiency as well as selectivity. Nitrogen-containing ligands, for instance, increase the nucleophilicity of CO<sub>2</sub> helping make its activation and conversion into useful products such as methanol or carbon monoxide easier. Computational studies with density functional theory (DFT) have shown that by fine-tuning the ligand environment this can substantially enhance the reaction mechanism through optimizing metals' coordination chemistry [8,9]. And so, the ongoing effort to find functional ligands is focused on systems which do not just hold up well under high performance conditions but where there is also sufficient stability and reactivity for practical CO<sub>2</sub> reduction applications that are industrial scalable [10].

Iron-quaterpyridine complexes have gained wide interest in the field of CO<sub>2</sub> reduction because they are effectively able to activate and subsequently convert CO<sub>2</sub> to important commodities, such as carbon monoxide and formic acid. These complexes utilize the coordination environment delivered by the quaterpyridine ligand for stabilizing the metal center and facilitating electron transfer during the reduction process. Iron, abundant and relatively cheap compared with noble metals like platinum and palladium, provides an attractive alternative in catalytic applications [11]. Although efficient, quaterpyridine complexes are generally overly complex in synthesis, insufficiently stable under reaction conditions, and difficult to tune in electronic properties for improving catalytic activity.

In response to these challenges, (1Z,1'Z)-N1,N1'-((Z)-ethene-1,2-diyl)bis(ethene-1,2-diamine) shortened as ZE-DiAm, a vinylic polyamine ligand, is introduced as a potential alternative. Besides being structurally simpler than quaterpyridines, ZE-DiAm is also

characterized by an ethene-1,2-diyl linkage that offers higher basicity and more flexible coordination behavior. These properties may enhance CO<sub>2</sub> binding and activation due to promotion of stronger interaction between the ligand and the metal center. In addition, the modular structure of ZE-DiAm may, possibly, allow easier tuning of catalytic properties, a very important endeavor in the optimization of CO<sub>2</sub> reduction processes. Departing from this background, computational studies were designed in this study to find the performance of ZE-DiAm to provide insights aimed at developing more efficient catalytic processes.

The aim of this study is to investigate the binding energy of CO<sub>2</sub> with [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup>, a novel iron(II) complex, using DFT. DFT has become a popular approach for the calculation of molecular interactions because it is capable of providing descriptions of electronic structure and properties of systems involved in bonding that are crucial in the design of catalysts [12]. This work is based on the notion that the higher basicity of the ZE-DiAm ligand could improve the stability of binding of CO<sub>2</sub> as compared to traditional iron-quaterpyridine complexes. Acetonitrile (MeCN) was employed as the L-type ligand to readily help the reaction catalytically since CO<sub>2</sub> reduction reactions are most often done in acetonitrile, which has good solubility for CO<sub>2</sub> and gives support to maintaining stability of the reactive species during the catalytic cycle [13,14]. These advantages, bring to the foreground that ZE-DiAm, with acetonitrile, may be a promising alternative as a design aid in the development of iron-based catalysts for reduction of CO<sub>2</sub>. This study is important in that it can provide such conceptual ideas that would later help one to design experiments for their validation. The computational results may expose such key interactions that enhance the catalyst's performance and hence be a further step toward more scalable CO<sub>2</sub> reduction technologies that are also commercially viable. In turn, this effort could make a contribution to the scale of climate change since the technologies could be used to convert CO<sub>2</sub> back into useful chemical feedstocks.

## 2. Experimental section

### 2.1 Geometry optimization

To optimize the geometry of the [Fe<sup>II</sup>(ZE-DiAm)(MeCN)(CO<sub>2</sub>)]<sup>2+</sup> complex, the initial structure was drawn using ChemDraw 22.0. The binding mode of CO<sub>2</sub> with the metal was drawn as η<sup>2</sup>-binding mode as this is the mode found to be likely happens in Fe(II) based complexes in previous computational study [15]. Following this, the structure was copied into ChemDraw 3D 22.0, where the optimization calculations were performed. In this software, the MM2 molecular mechanics algorithm was selected for the optimization process to minimize the potential energy of molecular systems as well as maintaining reasonable geometric parameters.

The optimization process involved minimizing the energy of the structure by adjusting iteratively bond lengths and angles to attain a stable conformation that is the most energetically favorable geometry for the complex. Thus, the optimized geometry could be a rather precise description of electronic and steric properties of [Fe<sup>II</sup>(ZE-DiAm)(MeCN)(CO<sub>2</sub>)]<sup>2+</sup>. The optimized structure of [Fe<sup>II</sup>(ZE-DiAm)(MeCN)(CO<sub>2</sub>)]<sup>2+</sup> was investigated, focusing on the bond lengths and angles. The values obtained help in better understanding of coordination setup and behavior of CO<sub>2</sub> around the iron center.

### 2.2 Computational methods

The experimental methodology used in this study included performing DFT calculations on the [Fe<sup>II</sup>(ZE-DiAm)(MeCN)]<sup>2+</sup> complex using Gauss software to determine its binding energy with CO<sub>2</sub>. The energy of the molecule was optimized after structuring using GaussView 6.0. This

optimization had to be done to minimize the total energy of the system, which helps model the molecular interactions accurately as in widely accepted practice in computational chemistry [16,17].

In the DFT calculations for the  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  complex, certain parameters were set. The hybrid functional chosen was B3LYP, which is noted for being an exchange-correlation hybrid functional that finds a balance between accuracy and ease of computation. Calculations were performed in the ground state, using a default spin configuration that includes a spin quintet state to properly represent the electronic structure of the iron center. The basis set used was 6-31G(d) with additional polarization functions to enhance precision in transition metal systems [18]. A charge of +2 was also assigned to the complex to make its ionic state description more accurate.

Equally, the  $\text{CO}_2$  molecule was built and optimized using the same DFT parameters mentioned earlier for geometry optimization. The set used included B3LYP method, ground state designation, default spin configuration, and a basis set 6-31G(d). The  $\text{CO}_2$  molecule was given a charge of 0 and a spin singlet state since it is non-radical and neutral. This optimization ensured that the computational model effectively represented the molecular characteristics of  $\text{CO}_2$ .

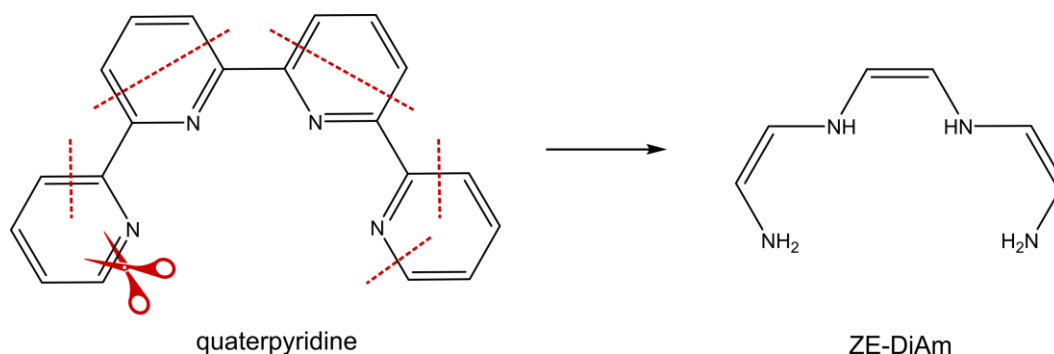
After the geometries of both the  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  complex and the  $\text{CO}_2$  molecule were optimized, the two components were combined to form the  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})(\text{MeCN})(\text{CO}_2)]^{2+}$  complex. The geometry of this new complex was also optimized to ensure accuracy. The binding energy of  $\text{CO}_2$  was calculated using the following formula:

$$E_{\text{binding}} = (E_{\text{Fe complex}} + E_{\text{CO}_2}) - E_{\text{Fe complex-CO}_2}$$

This equation [19] allowed the total energy of the Fe- $\text{CO}_2$  complex to be compared with the Fe complex and  $\text{CO}_2$  molecule energies to obtain an insight into the role of the ligand in  $\text{CO}_2$  activation. The energy values of the complexes were obtained from the output log files Gauss had generated. This approach corresponds to the common practice in computational chemistry, where DFT forms the standard method of investigation in the description of metal-ligand interactions as well as their catalytic activity.

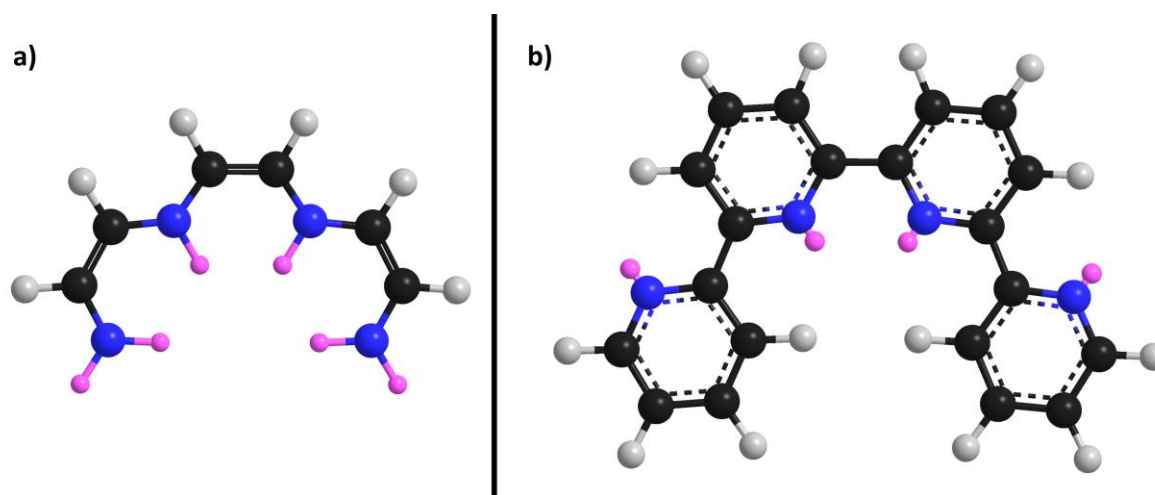
### 3. Results and discussion

ZE-DiAm was inspired by the simplification of the quaterpyridine ligand (**Scheme 1**), aiming to maintain the nitrogen atoms in the same positions while reducing complexity. By eliminating the aromatic moieties, the resulting ligand, ZE-DiAm, is predictably more basic than quaterpyridine. Energy minimization performed using ChemDraw 3D, as illustrated in **Fig. 1**, reveals that the rigid ethene-1,2-diyl backbone of ZE-DiAm fixes the positions of the nitrogen atoms, preventing free rotation. This rigidity enhances the stability of the Fe(II) complex, as the nitrogens remain optimally positioned for effective electron donation to the metal center. In contrast, quaterpyridine ligands allow the terminal pyridine nitrogens to rotate freely, often adopting configurations where the nitrogens are positioned opposite to those in the median pyridines. This flexibility reduces the efficiency of electron donation and may require additional energy to maintain the nitrogens in the correct positions during complex formation with Fe(II), ultimately affecting the overall stability of the complex [20,21].



**Scheme 1** Simplification of the quaterpyridine structure inspires the design of ZE-DiAm

In the optimized structure of  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})(\text{MeCN})(\text{CO}_2)]^{2+}$  (**Fig. 2**), some characteristics around the  $\text{CO}_2$  and  $\text{Fe}(\text{II})$  are found, they are, bond lengths and angles— $\text{C}=\text{O}$  at 1.2 Å,  $\text{Fe}-\text{N}$  at 1.9 Å,  $\text{Fe}-\text{O}$  at 1.9 Å, and  $\text{O}=\text{C}=\text{O}$  at  $142.9^\circ$ . The bond lengths are consistent with those found in similar transition metal complexes, confirming the realistic nature of the bonding interactions [22,23]. This effective coordination of ZE-DiAm's nitrogen atoms to the iron center is crucial for the complex's stability. A particularly noteworthy result is the ability of the  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  complex to bend the  $\text{O}=\text{C}=\text{O}$  bond angle from its typical linear configuration of  $180^\circ$  to  $142.9^\circ$ . This deviation indicates significant activation of the  $\text{CO}_2$  molecule, which is essential for its reduction. In its free form,  $\text{CO}_2$  is linear, and the bending of the molecule weakens its  $\pi$ -bonding, making it more reactive and susceptible to nucleophilic attack. This enhanced reactivity is crucial for the conversion of  $\text{CO}_2$  into the reduced species such as  $\text{CO}$  or  $\text{CH}_3\text{OH}$  [24]. In this sense, the results underline the potential of the complex  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  in  $\text{CO}_2$  reduction and denote its capacity as a catalyst in this major chemical transformation.

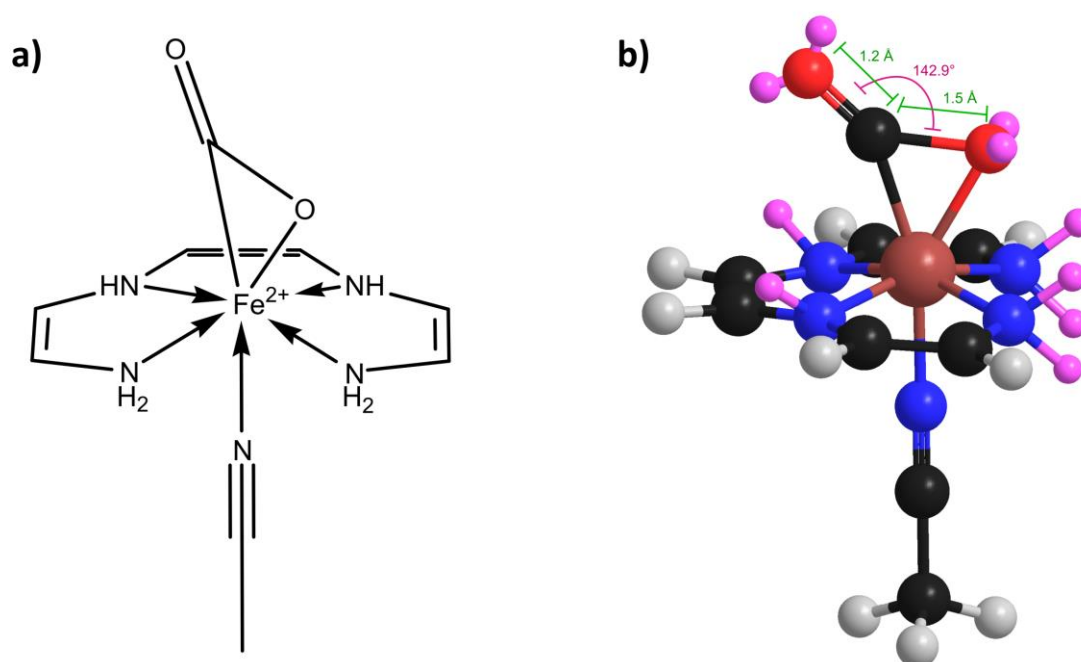


**Fig. 1** Geometry-optimized structures of a) ZE-DiAm and b) quaterpyridine, with hydrogen, carbon, nitrogen, and lone pairs depicted as gray, black, blue, and fuchsia spheres, respectively

The bending of  $\text{CO}_2$  to such an extent ( $142.9^\circ$ ) by  $[\text{Fe}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  reflects the strong interaction between the iron center and the  $\text{CO}_2$  molecule, indicating effective  $\pi$ -backdonation from the metal d-orbitals to the  $\pi^*$  antibonding orbitals of  $\text{CO}_2$ , weakening the  $\text{C}=\text{O}$  bonds. This

feature is common in transition metal complexes that facilitate CO<sub>2</sub> reduction. Such distortions of CO<sub>2</sub> geometry have been previously correlated with the ability of metal complexes to activate CO<sub>2</sub> for subsequent catalytic transformations. For example, iron porphyrin-based catalysts exhibit similar behaviors, where the CO<sub>2</sub> bond angle bends to 133.6° [25]. Comparatively, other iron complexes such as iron bipyridine and Fe(II)-quaterpyridine, which are widely studied for CO<sub>2</sub> reduction, typically bend the O=C=O angle to a lesser degree (around 160°), suggesting weaker activation and less efficient reduction capability [15,26].

The bond lengths within the [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup> complex further confirm its bent geometry and decode its affinity for reducing CO<sub>2</sub>. A Fe–C bond length of 1.9 Å and Fe–N bond lengths of 1.9 Å show fine coordination of the metal with the ligand, thereby stabilizing the complex for facilitation of electron transfer processes [27]. The bond length value is close to such reported highly active iron CO<sub>2</sub> reduction catalysts like iron tetraphenylporphyrins [27,28]. The C=O bond length in the complex is 1.2 Å (**Fig. 2 b**) which is slightly longer than the value for free CO<sub>2</sub> (~1.16 Å) [29], further showing interaction with the metal center which weakens the CO<sub>2</sub> bonds and thus makes it more reactive for reduction.



**Fig. 2** Geometry-optimized structure of [Fe<sup>II</sup>(ZE-DiAm)(MeCN)(CO<sub>2</sub>)]<sup>2+</sup> complex, a) 2-dimensional structure, b) 3-dimensional structure showing the bond length and angle around the CO<sub>2</sub> molecule, with hydrogen, carbon, nitrogen, oxygen, iron, and lone pairs depicted as gray, black, blue, red, reddish-brown, and fuchsia spheres, respectively

Thus, a considerably bent angle of CO<sub>2</sub> and the proper bond lengths between the iron center and the ligand in [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup> suggest that the complex may be a very effective catalyst for the reduction of CO<sub>2</sub>. These geometric parameters speak for the excellent electronic properties of the ZE-DiAm ligand, allowing not only for higher CO<sub>2</sub> binding energies but also for the molecular distortion that is needed to activate the CO<sub>2</sub> for reduction. With respect to all other analog systems known in the literature, this complex demonstrates an increased potential

for CO<sub>2</sub> activation, which makes it a potential candidate for further catalytic applications of CO<sub>2</sub> reduction.

The binding energy of CO<sub>2</sub> to [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup> was found to be 262.262 kJ/mol, significantly higher than those for other iron-based complexes of CO<sub>2</sub>, reported to lie between 33.890 and 125.520 kJ/mol [30,31]. This is an elevated binding energy value, denoting a strong electron donor strength of ZE-DiAm, which would environmentally stabilize the iron-CO<sub>2</sub> bond much better than quaterpyridine or bipyridine, well-studied ligands for their effects on CO<sub>2</sub> reduction catalysis [31]. With a binding energy value indicative of such a strong interaction between the iron atom and CO<sub>2</sub>, this may be functionally critical with respect to the reduction of CO<sub>2</sub>. Such a high value of binding energy not only signals good complex stabilization due to binding but also a real potential of activation toward CO<sub>2</sub>.

Higher binding energies were shown to be related positively with efficiency of CO<sub>2</sub> activation and successive reduction processes. A better binding of iron complexes with CO<sub>2</sub> was observed to enhance the catalytic performance in the reduction reaction of CO<sub>2</sub> [32]. Thus, the metal complex that can most effectively bend the O=C=O angle upon binding is predicted to have the best catalytic activity, as this distortion would lead to weakening of the C=O bonds and, thereby, promote the reduction pathway [15]. By this account, with binding energies higher than 200 kJ/mol, Fe-Ze-DiAm complexes can be viewed as potential candidates for CO<sub>2</sub> reduction due to their ability to stabilize the CO<sub>2</sub> molecule and lower the activation barrier. This stability and strong binding of CO<sub>2</sub> revealed in these calculations may enable the new proposed iron(II)-ZE-DiAm complex to stand under experimental conditions and raise turnover numbers and, hence, catalytic efficiency. One key factor contributing to this stability is the ligand's ability to donate electron density to the metal center, a property known as basicity. Understanding this aspect is crucial for further optimizing the catalytic behavior of the complex in CO<sub>2</sub> reduction pathways, and experimental studies are recommended to validate these computational predictions.

In coordination chemistry, basicity is one of the factors that determine the strength of binding between a metal and its ligand. Thus, it has an effect on the stability of the complex. More basic ligands are better electron donors, giving more electron density, which increases the metal-ligand bonding strength required for stability and performance of the metal complex under catalytic cycle [33,34]. The higher basicity of the vinylamine ligand ZE-DiAm rather than moderately basic quaterpyridine ligands is central to its ability to stabilize transition metals involved in CO<sub>2</sub> activation. This substituent may enhance the donation of electrons from nitrogen to the iron(II) center due to the presence of an alkyne-1,2-diyl linker in the polyamine backbone of ZE-DiAm. The vinylic amine groups—C=C—NH—present in ZE-DiAm relatively have greater donating strength compared with quaterpyridine ligands and thus enhance the overall basicity of the ligand. This increased basicity of the ligand also accounts for the strength of the coordinating bond between ZE-DiAm and the iron(II) center [35]. The increased basicity might be a major reason for the stability noted for the [Fe<sup>II</sup>(ZE-DiAm)MeCN]<sup>2+</sup> complex. These are the kinds of features that permit more sturdy interaction with CO<sub>2</sub>, a key factor in catalytic processes. Experimental efforts in the near future should confirm these above predictions by measuring the turnover frequency of freed catalyst under operation conditions, to test its stability and verify it as a promising high-performance catalysis.

The CO<sub>2</sub> emissions potentially targeted by this complex include the reduction of CO<sub>2</sub> to carbon monoxide (CO) concomitant with formation of other reduced products, such as formates and methanol. Additionally, more studies on putative transition states and intermediates that these pathways rely upon are required for potential future validation of such mechanisms

through experimental procedures. This study reveals that  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  to be an exceptionally potential catalyst in comparison with known iron-quaterpyridine complexes (higher binding energies and potential catalytic efficiency). This complex has several advantages compared to established systems like iron-quaterpyridine complexes, e.g. simplified structure and easier synthesis routes and functionalization possibility.

#### 4. Conclusion and future directions

Higher  $\text{CO}_2$  binding energies were calculated for the  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  complex (262.262 kJ/mol), as compared to previously reported iron-based  $\text{CO}_2$  reduction catalysts, indicate a stronger and more stable interaction with  $\text{CO}_2$ . Reorientation of the  $\text{CO}_2$  bond angle to  $142.9^\circ$  and optimized bonding distances around the iron center give indirect evidence for pronounced activation of  $\text{CO}_2$ , which is crucial in reduction processes. Altogether, this study reveals the potential of  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  as a new  $\text{CO}_2$  reduction catalyst that exhibits improvements in both stability and reactivity relative to traditional quaterpyridines. The more basic character of the ZE-DiAm ligand may also increase electron donation to form stronger metal-ligand interactions that are essential for catalysis. Experimental research may further verify the catalytic activity of this complex which would be suitable for potential applications in  $\text{CO}_2$  reduction technologies.

An experimental validation is necessary to evaluate the feasibility of  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  as an active catalyst in catalysis transitioning computational studies. Ideally proposed experimental setups should consider  $\text{CO}_2$  reduction conditions, the optimal amount of loading catalyst and deep product analysis that can be made with gas chromatography (GC) or NMR. Expected results will largely be centered on catalyst activity as typical with turnover number (TON) and turnover frequency (TOF), and product selectivity. The reason to interest and attract attention for  $\text{CO}_2$  reduction into valuable carbon species is the possibility of directly synthesizing chemicals like carbon monoxide, formic acid or methanol from abundant C1 feedstock instead than fossil fuels, offering a perfect paradigm to eliminate climate change by recycling atmospheric  $\text{CO}_2$ .

A route for the  $\text{CO}_2$ -to-elemental-carbon reduction is also interesting and important in energy storage as well as material synthesis in the future. To convert  $\text{CO}_2$  gas, however, is a series of transformations in which carbon monoxide and other possible intermediates form along with solid carbon. These pathways are complex and can be influenced to various degrees by factors such as temperature, pressure, the catalytic environment provided by the catalyst like  $[\text{Fe}^{\text{II}}(\text{ZE-DiAm})\text{MeCN}]^{2+}$  or similar catalysts. Here the electronic properties of the catalyst are crucial to mediate necessary electron transfers, while ligand interactions from ZE-DiAm should promote basicity and stabilize reaction intermediates in order to facilitate efficient electron transfer. The design of experimental setups should target surveying reaction conditions, product distribution analysis and elementally profiling the resulting carbon materials in terms on their morphology and crystalline nature. Expected results are efficiency, product yields and also some ideas on how to catalyze the desired transformation. The ripple effects of accomplishing that feat are enormous, from mitigating climate change to preserving dwindling natural resources. This transformative approach in the use of waste  $\text{CO}_2$  as a valuable resource to foster processes and technologies for carbon-neutralities by producing solid carbon, which is part of CCUS (Carbon Capture & Utilization Strategy) towards circular economy.



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