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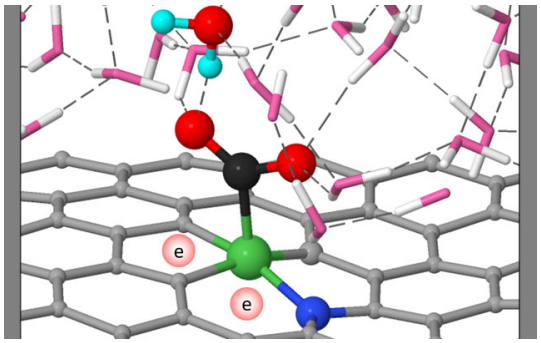
Supercomputer Simulations Help Advance Electrochemical Reaction Research

New process synthesizes valuable chemicals to use as electricity in lieu of fossil fuels

Single-atom catalysts have recently emerged as holding promise for solving environmental and energy issues. One such example, nickel embedded in graphene (a thin layer of graphite), has been shown to convert carbon dioxide, a molecule that causes the greenhouse effect, into carbon monoxide, which is an important feedstock for chemical engineering.



Electrochemical reduction of carbon dioxide catalyzed by a single nickel atom embedded in graphene with nitrogen dopant; "e" represents the electrons. Credit: Xunhua Zhao and Yuanyue Liu, UT-Austin

As a poisonous gas, carbon monoxide is often converted into carbon dioxide, such as in cars and trucks equipped with catalytic converters. This process is the reverse, which at first may sound a bit odd, but it provides an important role in synthesizing valuable chemicals for use as electricity in lieu of the Earth's rapidly depleting fossil fuels.

However, a better understanding of the atomic structure of this concoction is needed before nickel-embedded graphene can be used on a regular basis. To help with this challenge, researchers from the University of Texas at Austin (UT-Austin) recently simulated the catalytic mechanism and atomic structure of nickel-doped graphene using *Comet* at the San Diego Supercomputer Center (SDSC) and *Stampede2* at the Texas Advanced Computing Center (TACC). The simulations showed a clear picture of the catalyst's atomic structure so that researchers were able to better understand critical effects of surface change and hydrogen bonding, which were overlooked in previous models.

“Our work unveils the structure and mechanism underlying the performance of the catalyst - we found that the catalysis arises from the structure where the nickel atom is bonded with one nitrogen and three carbon atoms,” said Yuanyue Liu, an assistant professor of computational materials science at UT-Austin. “This structure can carry more charges than other hypothesized structures, and having more charges makes the electrochemical reaction faster, which means that the carbon dioxide can be converted to carbon monoxide with a higher rate.”

Liu further explained that this reaction can be facilitated by hydrogen bonding between water and the polar intermediate species generated during reaction, making it more favorable over other competing reactions that do not produce polar intermediate.

“This study not only explained a long-standing puzzle for an important catalyst but also highlighted the critical roles of change capacity and hydrogen bonding, which can help elucidate the mechanisms of other heterogeneous electrocatalysts in water to enable better design,” said Liu.

Published last month in the *Journal of the American Chemical Society,* Liu and co-author Xunhua Zhao outlined future plans to advance their studies on electrochemical reactions for energy conversion. Zhao, a postdoctoral student at UT-Austin, explained the importance of using supercomputers for this work.

“Our simulations required hundreds of CPUs running parallelly for tens of hours per calculation,” said Zhao. “We relied heavily on *Comet* and *Stampede2* to accurately model the complex processes of charge transfer and dynamically evolving hydrogen-bond networks.”

Specifically, Liu and Zhao used a molecular dynamics simulation technique based on quantum mechanics – an examination of molecular structures. They are now focused on collaborations with experimentalists to test their computational results.

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