

Original Article: Exploring Novel Catalysts for Enhanced CO₂ Reduction Reactions

Mohammad Javad Gandomi

Department of Chemical Engineering, Hakim Sabzevari University, Sabzevar, Iran



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ABSTRACT

The excessive use of fossil fuels to carry out industrial activities by humans has led to the excessive emission of CO₂ in the atmosphere. According to the latest reports, the concentration of carbon dioxide has exceeded 400 ppm. A concentration of 400 ppm means that for every one million particles of air, there are 400 particles of carbon dioxide molecules (0.04%). This extremely high concentration of CO₂ has led to global warming and many negative consequences for the Earth's climate system, but despite the harm and dangers caused by carbon dioxide, this gas can be a useful source of carbon for the synthesis of valuable chemicals and fuels. Therefore, a team of researchers recently selected a highly efficient and stable copper-based catalyst (DFNS/TiO₂-Cu) to convert CO₂ to CO. In addition to providing researchers with information on the regulation of strong metal interactions, this study shows that this catalyst can remain stable for more than 200 hours. There are many reports of noble metal catalysts, but their application is limited due to the moderate catalytic performance and high cost of these metals. In the family of non-noble metal catalysts, copper-based catalysts are among the most widely used catalysts that have a good capacity to perform industrial processes, but unfortunately, the low Tamman temperature of copper limits its activity and long-term stability. As a result of the research, scientists introduced a catalyst with active copper sites on titanium oxide-coated Nano fibrous silica (DFNS/TiO₂-Cu) to convert CO₂ to CO.

*Corresponding Author: **Mohammad Javad Gandomi** (Mjg1996@yahoo.com)

Introduction

The result of tens of years of photosynthesis and hundreds of thousands of years of pressure and heat caused by geological activities is the conversion of carbon dioxide in the earth's atmosphere into long chains of hydrocarbons that make fossil fuels, but humans cannot convert the excess carbon dioxide in the atmosphere into useful hydrocarbons. Millions of years to wait. However, advances in chemistry can help us speed up part of this process. Recent achievements in the field of making catalysts that can convert carbon dioxide into carbonaceous molecules can promise a promising perspective in the field of collecting harmful pollutants from the earth's atmosphere and turning it into usable materials [1].

Although the hydrocarbons produced by the new catalysts are of short chain type, this process is more efficient than all previous methods in converting carbon dioxide into hydrocarbons. Carbon dioxide, known to be a major factor in global warming, will become a new space if a small American company succeeds in turning it into gasoline [2].

At first glance, this company seems as crazy as the medieval alchemist Nicolas Flamel, except that it is no longer about turning lead into gold, but rather polluting into clean energy. While the Obama administration has prioritized the fight against global warming and the search for clean energy, the company carbon Science, which has registered its discovery, hopes to mobilize politicians and opinion to be the first to this bet succeeds.

The small company in Santa Barbara, California, says it is ready to build its first operational-scale pilot plant that could begin producing next-generation biofuels by the end of 2010. Its chief operating officer, Byron Elton,

explained that all he had to do was find a partner to launch the project.

During a meeting in New York, he said: Our partner can be someone who produces a lot of CO₂. If a partnership closes within nine months, this new type of biofuel could begin production by the end of 2010, Elton says, while acknowledging that the timeline could be ambitious [3].

In fact, the fibrous morphology and high surface area of DFNS/TiO₂ cause better dispersion and high loading of active sites of copper nanoparticles, and from these findings, it was found that this catalyst has excellent catalytic performance for CO₂ reduction with an efficiency of 5350 mmol g⁻¹ h⁻¹. It has performed better than all copper-based thermal catalysts. It was also found that the conversion of CO₂ to CO follows a path in the redox reaction with the help of hydrogen.

The technology developed by carbon science uses microorganisms, which it calls biocatalysts. It is necessary to destabilize the carbon dioxide by mixing it with water. Then the microorganisms protected by specially produced polymer shells are responsible for the recombination of hydrogen and carbon to produce hydrocarbons.

This mechanism is the same one that was implemented in the creation of hydrocarbons in nature, but in the process of carbon science, biocatalysts are protected and reused. Therefore, gasoline can be produced at a very competitive cost price [3].

Carbon Sciences, a listed company that employs just eight people, claims to be the most advanced in this research, but it's not the only one exploring this avenue. Investigative researcher Craig Venter, whose team first announced that it had successfully sequenced the human genome in 2000, announced in February 2008 that he thought it would be possible to produce biofuel within 18 months. The fourth generation is, they say, not based on

agriculture like ethanol, but based on carbon dioxide [4].

Today, the J. Craig Venter Institute mainly praises advances in the use of algae to break down and recombine CO₂ into hydrocarbons. These initiatives have attracted the attention of American officials. All of them are more popular in the United States. Because this country supplies half of its electricity from coal-fired power plants, which emit a lot of carbon dioxide.

Therefore, its ecological cost is criticized. "The question is not if we use coal, but how we use it," influential Democratic Sen. Byron Dorgan said earlier this month. He said: In the economic improvement plan that was voted at the beginning of the year, 3.4 billion dollars have been budgeted for this purpose. According to Byron Elton, a windbreaker that carbon scientists like to use, the technology, developed by chemical engineers at Stanford university, combines excess carbon dioxide with just the right amount of hydrogen to form a chain of ethane, propane, and even butane molecules [5].

They form Molecules that can become the source of production of new generation fuels. In recent years, efforts have been made to invent a cost-effective way to extract carbon dioxide from the atmosphere and convert it into useful materials such as fuel or plastic. Materials that people are willing to spend money on. The challenge that scientists are facing is to invent a method that will convert at least a small part of the billions of tons of carbon dioxide that enters the earth's atmosphere into useful materials in different ways.

For this method to be economical, the maximum possible amount of carbon dioxide in the combustion gases should be collected and converted into the desired materials through a quick and cheap process. Carnillo says: In order to trap the maximum possible amount of

carbon dioxide, the hydrocarbon chain formed must be as large as possible, but using current technologies, it is not possible to produce long chains of hydrocarbons.

A lot of heat and pressure is needed to form long hydrocarbon chains. Therefore, the efficiency of the process decreases and the costs skyrocket. Carnillo and his research team focused on organic polymers. Materials with many pores that can be changed in size to create a suitable structure for trapping carbon molecules and combining it with hydrogen and finally forming a hydrocarbon chain. In this process, another metal catalyst is used to speed up the process [6].

This material is a combination of ruthenium element with titanium oxide. Researchers can significantly increase the efficiency of the process by modeling the pores of polymeric structures. The result was extraordinary. A 10-fold increase in the molecular mass of carbon chains, as well as the production rate of four-carbon butane chains using catalysts covered with organic polymers has increased up to 1000 times.

"Catalysts without polymer coating absorb a lot of hydrogen on their surface," says Cheng Shuang Zhou, a PhD student working in Carnillo's lab. As a result, it reduces the ability of the material to trap carbon molecules and combine it with hydrogen, but when the catalyst surface is covered with porous polymers, the absorption rate of carbon and hydrogen molecules can be balanced and longer chains of carbon can be produced in the same reactions [7].

Ideally, any amount of carbon that can be extracted from the Earth's atmosphere should be safely contained, either in the form of fuel and other consumables or carbon-based materials for which there is no specific use. Production of fuels with zero net carbon emissions can economically justify the process of carbon dioxide extraction and fuel

production, and on the other hand, help control greenhouse gas emissions [8].

Butane is heavier than other hydrocarbons and is not as likely to evaporate at room temperature and leak into the atmosphere as other short chain hydrocarbons. Therefore, the production of such a hydrocarbon is considered an important achievement for researchers. However, with more research, this chemical process can be used to produce more stable types of hydrocarbons. Materials that can be the basis for the production of durable and useful structures for the safe storage of carbon molecules [9].

Of course, it is clear that simulating nature's method of collecting carbon and using it in biological structures and inventing a similar process is the first priority of researchers. What could be better than converting carbon dioxide into a variety of plants and tree leaves for more photosynthesis?

But if continuing current methods will reduce our dependence on fossil fuels in the future, investing in this technology will be justified. The latest development in this field is a new catalyst that can remove short-chain carbon molecules made from carbon dioxide at a faster rate than previous methods [10].



Figure 1. The new technology, developed by chemical engineers at Stanford University, converts waste carbon dioxide and a good dose of hydrogen into chains of ethane, propane and even butane. These are all molecules that can act as a fuel source.

"We can essentially produce gasoline," says Stanford University chemical engineer Matteo Cargenello. In recent years, no effort has been spared to find economical ways to take carbon out of the air and turn it into something people want to pay for, like fuel or synthetic materials like plastic [11].

The serious challenge is to reduce the enormous amounts of carbon we pump into the atmosphere every year. In other words, we need a fast process that can be done cheaply and remove as much carbon in each molecule as possible. "To capture as much carbon as possible, we want the longest chain hydrocarbons," says Cargenello. Chains with 8 to 12 carbon atoms would be ideal. Current technologies strive to approach this goal. The longer the chain, the more heat and pressure is required, making the process less efficient and more expensive.

Cargenello and his team focused their research on organic polymers. Materials with pores that can be easily scaled to suit the type of structure needed to maximize the reactivity of carbon dioxide and hydrogen in chains. In combination with an effective catalytic metal to speed up the process, in this case, the element ruthenium combined with titanium oxide, researchers can improve efficiency by modeling the polymer's pore structures [12].

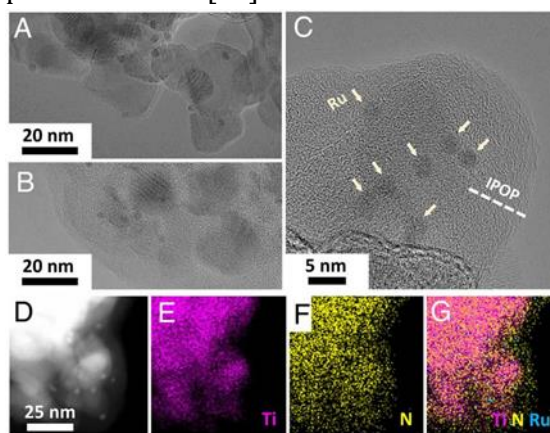


Figure 2. The researchers improved the efficiency and showed a 10-fold increase in the turnover of high molecular weight carbon

chains. Remarkably, the production rate of four-carbon butane chains increased 1000 times by coating the catalyst in a special type of porous organic polymer.

"An uncoated catalyst is covered with a lot of hydrogen on its surface, limiting the carbon's ability to find other carbons to bond with," says Cheng Shuang Zhou, a chemical engineer and doctoral student in Cargenello's lab. The porous polymer controls the ratio of carbon to hydrogen, allowing us to create longer carbon chains from similar reactions. Ideally, any carbon we can pull out of the atmosphere should be retained [13].

Carbon-neutral fuels may help pay for such technologies at the cost of creating a cycle for atmospheric carbon. The step of producing more butane, which is less likely to quickly vaporize to gas at room temperature and leak into the atmosphere like smaller chains, is critical. However, with further research this type of chemical process may produce more stable forms of hydrocarbons that can be combined into more stable structures that we want to safely store.

Of course, nature's solution to extract carbon and use it in biological materials will always be the number one priority. There's nothing better than keeping carbon dioxide in the form of plants, but if it helps us get off our dependence on fossil fuels sooner, it's a solution worth pursuing. In this technology, a new catalyst made of organic polymers has been used, which, along with a metal catalyst made of a combination of titanium oxide and ruthenium, significantly increased the efficiency of the process of converting carbon dioxide into fuel. In preliminary simulations, it was determined that this catalyst is able to increase the conversion of carbon dioxide to high molecular weight carbon chains by 10 times.

Also, by covering the metal catalyst with organic polymers, the production rate of butane chains with four carbon atoms

increased by a thousand times. According to the researchers, metal metal catalysts keep large amounts of hydrogen atoms on their surface, which reduces their efficiency in connecting carbon atoms together, but covering these catalysts with porous polymers controls the rate of connecting carbon and hydrogen atoms on the catalyst surface.

And enables the formation of longer carbon chains. Producing fuel from carbon dioxide is an effective way to reduce the amount of this greenhouse gas in the atmosphere and also to reduce the consumption of fossil fuels. In May of last year, the Mona Loa Observatory recorded 415.26 parts per million for carbon in the atmosphere, which was the highest level of carbon dioxide recorded until then, but now, after 12 months, the new record is 416. 321 parts per million have been recorded. According to the research of the US National Oceanic and Atmospheric Administration using the polar ice layers, this amount of atmospheric carbon dioxide is unprecedented in the last 800 thousand years, and the reduction of human activities due to the corona pandemic has not affected this increasing trend [14].

For this reason, the development of methods to convert this gas into fuel is of interest to researchers all over the world. In the quest for cleaner air and a healthier environment, industries are grappling with the challenge of reducing harmful greenhouse gas emissions. Meanwhile, nitrogen oxides (NO_x) are a significant threat, contributing to smog, acid rain, and respiratory problems.

Fortunately, innovative technologies such as selective catalytic reduction (SCR) offer a powerful solution. At the heart of this process lies the SCR catalyst, a critical component that facilitates the conversion of NO_x into harmless nitrogen and water vapor. SCR catalysts are the unsung heroes of NO_x reduction [15].

These complex structures, often composed of ceramic materials such as titanium oxide, are

injected with active catalytic components such as base metal oxides (vanadium, molybdenum) or precious metals. Their porous design provides a large surface area where the magic happens. NO_x molecules in the exhaust gas react with a reductant, usually ammonia or urea, in the presence of a catalyst. This reaction converts NO_x into harmless nitrogen and water, effectively cleaning the exhaust gas before it is released into the atmosphere.

The specific reactions involved depend on the reductant used:

With ammonia: $4\text{NO} + 4\text{NH}_3 + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O}$

With urea: $2\text{NO}_x + (\text{NH}_2)_2\text{CO} + \frac{1}{2} \text{O}_2 \rightarrow 2\text{N}_2 + \text{CO}_2 + \text{H}_2\text{O}$

These reactions occur within a specific temperature window, typically between 357 and 447°C, although lower temperatures can be effective with longer retention times [16].

Choosing the right SCR catalyst

SCR catalysts come in many forms, each with their own strengths and weaknesses, making the selection process critical for optimal performance. Honeycomb catalysts, similar to a honeycomb structure, are the most common type. Their advantages are:

1- High surface area: This allows more contact between the exhaust gas and the catalyst and reduces NO_x efficiently.

2- Good mixing: The honeycomb structure facilitates the turbulence and ensures the complete mixing of exhaust gas and reducer. However, they are also prone to clogging, as fly ash and other suspended particles can collect in the ducts, reducing efficiency and increasing pressure drop.

3- Greater pressure drop: Complex channels can restrict gas flow and require more energy to push the exhaust through the system.

Plate catalysts

Plate catalysts consist of flat, parallel plates that are covered with catalyst material.

A) The advantages of sheet catalysts are:

1- Lower pressure drop: The open structure allows for easier gas flow and lower energy consumption.

2- less prone to clogging: smooth surfaces trap less suspended particles.

B) Disadvantages of sheet catalysts are:

1- Lower surface area: Compared to honeycomb catalysts, they offer less contact surface area for reaction and potentially reduce efficiency.

2- Larger size: plate catalysts need more space, which can be a limitation in some applications [17].

Corrugated catalysts

Corrugated catalysts offer a balance between honeycomb and plate types. With a wave structure that provides a higher surface area than plate catalysts while maintaining a lower pressure drop than honeycomb catalysts. However, they are less common and may cost more [18].

The role of regenerators in SCR

The effectiveness of SCR technology also depends on the choice of regenerator, each of which has its own safety and management considerations.

1- Anhydrous ammonia: very efficient and widely used in large industrial applications. However, it is a dangerous gas that requires strict safety protocols for storage and handling.

2- Aqueous ammonia: a safer alternative to anhydrous ammonia, easier to store and transport. However, it needs to be vaporized before being injected into the SCR system.

3- Urea: the safest option that is usually used in car applications. It requires thermal decomposition to convert to ammonia before

reacting with NO_x. However, this process adds complexity to the system.

SCR catalysts in action

With a variety of catalyst and regenerator options, SCR technology finds wide application in various industries struggling with NO_x emissions [19].

1- Power plants: SCR systems are necessary to reduce NO_x emissions from coal, oil and gas power plants. They are usually installed between the economizer and air preheater, and ammonia or urea is injected upstream of the catalyst.

2- Heavy-duty vehicles: SCR catalysts are critical to meet strict emission standards for diesel engines in trucks, buses, and other heavy-duty vehicles.

3- Marine engines: SCR systems help reduce NO_x emissions from ships and contribute to cleaner air in ports and coastal areas.

4- Industrial boilers and furnaces: SCR technology is used in various industrial processes such as cement production, waste incineration and chemical production to control NO_x emissions [20].

Challenges and solutions in SCR technology

Despite its effectiveness, SCR technology faces challenges that require careful consideration and mitigation strategies.

1- Catalyst poisoning: Sulfur compounds, heavy metals and other pollutants can deactivate the catalyst and reduce its efficiency and lifespan. Proper fuel selection, fuel cleaning, and use of upstream scrubbers can help prevent poisoning [21].

2- Catalyst binding: fly ash and suspended particles can accumulate in the catalyst. Especially in types of honeycombs that lead to an increase in pressure drop and a decrease in performance.

3- Ammonia slippage: Unreacted ammonia that passes through the SCR system can

contribute to greenhouse gas emissions and form ammonium salts that can damage downstream equipment. Optimizing ammonia injection and ensuring proper mixing can minimize slippage.

4- Cost: The implementation and maintenance of SCR systems can be expensive and requires a detailed cost-benefit analysis. SCR catalysts are at the forefront of the fight for cleaner air. By understanding their functions, types and applications, industries can harness the power of this technology to minimize their environmental footprint and contribute to a healthier planet [22].

To reduce the amount of carbon dioxide gas, it can be converted into useful chemicals through reactions. One of these reactions is the photogas-water process (RWGS). In this process, carbon dioxide gas is combined with hydrogen gas and carbon monoxide gas and water are produced. Carbon dioxide gas is converted into various chemical and fuel substances with hydrogen gas in various processes and with the presence of various catalysts. Therefore, it is very important to prepare the right catalyst for this reaction.

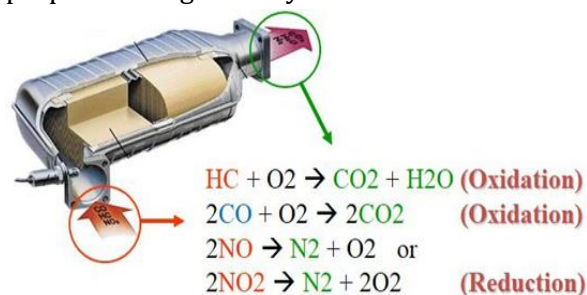


Figure 3. Oxidation and reduction inside the reactor

The reason for choosing the three-way name for these converters is to simultaneously reduce three types of toxic gases and air pollutants including carbon monoxide (CO), hydrocarbons from incomplete combustion (HC) and nitrogen oxide (NO_x) and convert them into non-toxic gases of water vapor H₂O, nitrogen N₂ and carbon dioxide CO₂ are used.

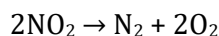
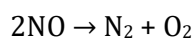
The catalytic converter uses two types of catalysts to purify pollutants:

1- Reduction Catalyst.

2- Oxidation Catalyst.

Both types of catalysts consist of a ceramic structure with a metal coating, usually platinum, rhodium, and palladium.

1- Pollution reduction catalyst: The function of the reduction catalyst is the first stage of refining by catalytic converters. This type of catalyst reduces NOX nitrogen oxides by using platinum and rhodium. When NO or NO₂ molecules hit the catalyst, they lose their nitrogen. The available nitrogen is absorbed by the catalyst and the remaining oxygen is released from the catalyst in the form of O₂. Nitrogen absorbed by the catalyst combines with other nitrogen molecules in the catalyst and becomes N₂ molecule [23].



2- Oxidizing catalyst: The function of the oxidizing catalyst is the second stage of refining by catalytic converters. This type of catalyst reduces the amounts of unburned hydrocarbons and carbon monoxide by burning (oxidizing) them by passing them through platinum and palladium catalyst plates.

Substrate carrying reagents

Substrate-carrying reactants, a honeycomb assembly consisting of a large number of cells. The purpose of the specific geometry is to achieve a large surface and absorb more species by the coating and as a result more reaction [24].

Types of carriers

1- Metal reagents: metal carrier reagents have more active surface. They create less pressure drop. They have better mechanical, thermal and heat transfer resistance. On the other hand, due to the complex technology, they are more difficult to make and therefore more expensive, and their weight is more compared to ceramic bearings.

2- Ceramic reactors:

Wash coat coating

The cover is a thin layer that is covered on the inner surface of the bees' nest. Their materials include alumina (Al₂O₃), zirconia (ZrO₂), ceria (CeO₂).

- ✓ A suitable substrate for depositing catalytic metals.
- ✓ Increased surface area for species adsorption due to porous structure.
- ✓ Increased thermal resistance and stability [25].

Gold nanoparticles

Gold nanoparticles can act as a catalyst and convert carbon monoxide into carbon dioxide. Gold nanoparticles as Nano catalysts in removing pollutants or cleaning car exhaust gases increase the intensity of desired chemical reactions. The properties of gold nanoparticles are very different from bulk gold. Only when gold atoms are confined in nanometer sizes do they exhibit catalytic properties and behavior.

The rare property of gold nanoparticles

Their activity is at low temperatures (ambient temperature and even lower), which is their advantage, which significantly reduces the operating costs of chemical units and increases the selectivity of the reaction [26].

The important feature of gold-based catalysts is the oxidation of carbon monoxide at low temperatures, and these types of catalysts are active even at minus 70^oc.

Another characteristic of nanoparticles is the high surface-to-volume ratio in this group of materials and particles.

The selectivity or selection of materials is one of the most important properties and characteristics of catalysts. Gold nanoparticles are one of the catalysts that have been proposed for the removal of carbon monoxide due to their proper selectivity and favorable activity [27].

Conclusion

Now, researchers at MIT and elsewhere have developed an entirely new type of catalyst material called a metal hydroxide organic framework (MHO), which is made of inexpensive and abundant components. The family of materials allows engineers to precisely tailor the structure and composition of the catalyst to the needs of a specific chemical process, and can then match or exceed the performance of conventional, more expensive catalysts. The findings are described in a February 24, 2022, issue of the journal *Natural Materials* in a paper by MIT postdoctoral fellow Shuai Yuan, graduate student Jiayu Peng, professor Yang Shao-Horn, professor Yuri Roman-Leskov, and nine others. Oxygen evolution reactions are one of the common reactions in the electrochemical production of fuels, chemicals and materials. These processes include the production of hydrogen as a byproduct of oxygen evolution, which can be used directly as fuel or undergo chemical reactions to produce other transportation fuels, ammonia production, use as fertilizer or chemical raw materials, and carbon dioxide reduction for control the emission of greenhouse gases. But without help, these reactions are slow, Shao-Horn says. You have to sacrifice voltage, or energy, to speed up the reaction.

Due to the additional energy required, the overall efficiency is low. "That's why people use

catalysts," he says. Because these materials naturally increase the reactions by reducing the input energy. But to date, these catalysts have all relied on expensive materials or delayed transition metals, which are extremely rare, says Roman-Leskov.

For example, iridium oxide, and there has been a lot of effort in the community to find alternatives based on earth-abundant materials that have similar performance in terms of activity and stability. The team says they have found materials that provide exactly the right combination of properties.

Roman-Leskov says other teams have explored using metal hydroxides such as nickel and iron hydroxides, but such materials have been difficult to adapt to the requirements of specific applications. However, the reason our work is so exciting and almost relevant is that we have found a way to match the properties to the nanostructure of these metal hydroxides in a unique way.

The team borrowed from research done on a class of compounds known as metal-organic frameworks (MOFs), a type of crystalline structure made of metal oxide nodes linked together by organic bonding agent molecules. By replacing the metal oxide in such materials with specific metal hydroxides, the team discovered that it was possible to create finely tuned materials that had the stability needed to be potentially useful as catalysts.

"You put these chains next to each other, and they actually drive the formation of metal hydroxide sheets that are connected to each other, and then they stack up and are more stable," says Roman-Leskov.

This has several advantages, he says. Through precise control over the patterning of the nanostructure, allowing precise control of the electronic properties of the metal as well as providing greater stability that enables them to withstand long periods of use. In testing such materials, the researchers found the catalyst's

performance surprising, Shaw-Horn says. This method is comparable to the advanced oxide method that catalyzes the oxygen evolution reaction.

The materials, which are composed mainly of nickel and iron, should be at least 100 times cheaper than existing catalysts, they say, although the team has not yet done a full economic analysis. "This family of materials really offers a new space for tailoring active sites for catalyzing water splitting to produce hydrogen with reduced energy input, to the precise needs of any given chemical process where such catalysts are needed," Shaw-Horn says. To fulfill Peng says: simply by substituting different metals instead of nickel in the composition, these materials can be five times more tunable than existing nickel-based catalysts.

This work potentially offers many relevant avenues for future exploration. So far, the materials have been tested in small-scale laboratory devices, and the team is currently exploring the issues of trying to scale the process to commercially relevant scales, which could still take several years, but Shao-Horn says the idea.

It has great potential to help accelerate the production of clean, emission-free hydrogen fuel, so that we can reduce the cost of hydrogen from the process, while not being limited by the availability of precious metals. This is important. Because we need hydrogen production technologies that can scale.

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