CARBON DIOXIDE FIXATION
THE MUST AND THE PATH

BY

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ABSTRACT

Abnormal climate change and uncontrolled natural calamities and catastrophes are increasing in the recent years. Reduction of carbon dioxide level in the atmosphere is an immediate need as CO₂ plays a vital role in the enhanced greenhouse effect leading to climate change. The CO₂ concentration in the atmosphere is increased due to the continued imbalance of its production and consumption. The reduction of CO₂ is significantly affected by deforestation. Natural fixation of CO₂ in the atmosphere such as photosynthesis and ocean carbon cycle is not enough to control the growing amount of CO₂. Artificial fixation of CO₂ is mandated. Carbon dioxide is captured and extracted by various methods. Techniques using effective absorber, conventional and nanocatalytic methods, capturing using enzymes and capturing using microalgae methods are few pathways to reduce CO₂. The turnover number of CO₂ capture in the catalytic process is directly proportional to the surface area of the catalyst material and the nature of the active site of the catalyst. CO₂ can be converted into useful chemicals such as methane, methanol, oxygen and biomass. The continued challenge of converting CO₂ into useful material and the need for the large CO₂ reduction using a little amount of reducing material is a growing research.

Keywords: Carbon dioxide, global warming, enhanced greenhouse effect, carbon dioxide balance, carbon dioxide capture, natural and artificial fixation of CO₂, nanocatalysts, enzyme and microalgae.

INTRODUCTION

The overwhelming majority of scientists agree that our globe is undergoing major climate change. They also agree that the level of carbon dioxide in the atmosphere is rising significantly. CO₂ is considered to be one of the main contributors to the greenhouse effect, causing significant impact on climate change, increasing precipitation, developing storm patterns and increased tide heights and sea levels (1). The main source of atmospheric CO₂ originates from human impact on the environmental emissions as a consequence of the consumption of fossil fuels. The CO₂ capture process started in the 1970’s due to the interest in using CO₂ in the process of enhanced oil recovery. Since then, the awareness of the greenhouse effect emerged and the techniques for CO₂ - capture had been attempted. Typical processes for CO₂-capture include gas-solid adsorption, gas-liquid absorption, cryogenic techniques and membrane systems. Among these, the gas-liquid absorption processes have been extensively studied and are currently considered the most effective and relatively low cost method for reducing CO₂ emissions from fossil fuel fired power plants. The majority of the absorption processes rely on a specific solvent to either react or physically absorb the CO₂ in the flue gases. Reacting solvents rely on the characteristics to weakly bond with CO₂ to form intermediate compounds followed by recovering the original solvent by applying heat.
GLOBAL WARMING

There is a gradual increase in the average temperature of the Earth’s atmosphere in the last 100 years. It has risen about 1°C since 1900; though there is no scientific evidence to suggest that global average temperatures will remain constant or decline in the next 100 years. Recent scientific reports conclude there is a 40% chance that warming will exceed this range and only a 5% chance that it will be less (2). As land is more responsive to atmospheric temperature changes than the oceans, the temperature increase will be greater over the continents than in the oceans. As the main absorption frequency associated with CO₂ that deals with the greenhouse effect is at 15 micrometers and that is the peak of the planets blackbody emissions curve, adding more CO₂ into the atmosphere has a profound effect especially when feedbacks are taken into account (3). Most radiation in the atmosphere above 4µm comes from black-body radiation emitted by objects heated up by absorbing shorter wavelength radiation. Greenhouse gases have the ability to absorb energy at these longer wavelengths, specifically between 4µm and 100µm. Though absorption of higher energy modes is possible, it is largely inconsequential to the greenhouse effect as those frequencies are not emitted by the earth's surface.

GREENHOUSE EFFECT

Greenhouse gas (abbreviated as GHG) is a gas in an atmosphere that absorbs and emits radiation within the thermal infrared range. Greenhouse effect is one of earth’s natural processes to regulate the temperature of our planet. It is essential for life on earth and is unquestionably real. It is a result of heat absorption by greenhouse gases in the lower atmosphere and re-radiation downward of some of that heat. Greenhouse gases greatly affect the temperature of the Earth. Without a natural greenhouse effect, the temperature of the earth would be about 0°F (-18°C) which is colder than the present 57°F (14°C) suitable for human life survival. In the 19th century, scientists realized that gases in the atmosphere cause a "greenhouse effect" which affects the planet's temperature. These scientists were interested primarily in the possibility that a lower level of carbon dioxide gas might explain the ice ages of the distant past. The primary greenhouse gases in the Earth's atmosphere are water vapor, carbon dioxide, methane, nitrous oxide and ozone. Since the beginning of the Industrial revolution, the burning of fossil fuels has contributed to the increase in carbon dioxide in the atmosphere from 280ppm to 390ppm. The emissions of carbon dioxide are directly proportional to energy consumption.

ENHANCED GREENHOUSE EFFECT

The concern is not with the fact that we have a greenhouse effect, but whether human activities are leading to an enhancement of the greenhouse effect by the emission of more and more greenhouse gases through fossil fuel combustion and deforestation (4). Carbon dioxide from combustion of coal, oil and gas concentration is increasing at a rate of 1.9 ppm/year since 2000. The pre-industrial level of CO₂ was about 280 ppm and the current level is about 380 ppm. Intergovernmental Panel on Climate Change (IPCC) projects the growth range from 490 ppm to 1260 ppm by the end of 21st century figure 1.
GREENHOUSE GASES AND THEIR CONTRIBUTIONS

There major greenhouse gases and their contributions are illustrated in Table 1.

<table>
<thead>
<tr>
<th>Green House Gases</th>
<th>Greenhouse Effect (%)</th>
<th>Natural (%)</th>
<th>Man Made (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water Vapor</td>
<td>95.00</td>
<td>94.999</td>
<td>0.001</td>
</tr>
<tr>
<td>Carbon Dioxide (CO₂)</td>
<td>3.618</td>
<td>3.592</td>
<td>0.117</td>
</tr>
<tr>
<td>Methane (CH₄)</td>
<td>0.360</td>
<td>0.294</td>
<td>0.066</td>
</tr>
<tr>
<td>Nitrous Oxide (N₂O)</td>
<td>0.950</td>
<td>0.903</td>
<td>0.047</td>
</tr>
<tr>
<td>Misc. gases (CFC’s, etc.)</td>
<td>0.072</td>
<td>0.025</td>
<td>0.047</td>
</tr>
<tr>
<td>Total</td>
<td>100.00</td>
<td>99.72</td>
<td>0.28</td>
</tr>
</tbody>
</table>

Table 1: Greenhouse gases and their contributions

The effect of greenhouse gas not only depends on the quantity but also on their chemical nature. For example, the influence of methane and CFC are very high though they are present in lower quantities. Though water vapor dominates the greenhouse effect among the contributing gases, CO₂ plays a major role as it influences the increase of water vapor concentrations. The concentration of nitrous oxide began to rise at the beginning of the industrial revolution. Miscellaneous greenhouse gases such as CFC’s are mostly synthesized by the use of refrigerants, aerosol propellants and cleaning solvents.
CARBON DIOXIDE BALANCE

Approximately 600 bt of carbon moves back and forth between the atmosphere and the oceans, and between the atmosphere and the land biosphere. Although these exchange rates are large relative to the total amount of carbon stored in the atmosphere, the concentration of 2200 bt CO₂ was constant. Marine plants and animals play a role in the uptake and release of carbon dioxide in the ocean (5). Atmospheric concentrations of CO₂ were constant because the carbon being removed from the atmosphere in some places exactly matched the CO₂ being added to the atmosphere in other places figure 2.

Most of the CO₂ released from the burning of fossil fuels and other human activities is stored either in the atmosphere or in the oceans. The CO₂ that remains in the atmosphere acts as a greenhouse gas, absorbing long-wavelength radiation (heat) in the atmosphere. CO₂ taken up by the oceans does not affect the Earth's heat balance. So, an understanding of the air-sea exchange of CO₂ is an essential part of understanding the Earth's climate system and the potential impact of future CO₂ emissions.

WHY TO REDUCE CO₂?

The present society intends on achieving influences and conveniences has forgotten how precious nature is. Enhanced CO₂ is a very big challenge we are faced with today. The impact of CO₂ on human health can be of different extend starting from minor shortness of breath to fatal death. Figure 3, shows the effect of CO₂ concentration in breathing dose.
It is very clear that CO₂ can cause damage to health and also responsible for enhanced greenhouse effect leading to global warming. Thus identifying the ways to reduce CO₂ becomes critical.

**WAYS TO REDUCE CO₂**

The main source of atmospheric CO₂ originates from anthropogenic emissions as a consequence of the consumption of fossil fuels. The continuous production and depletion of CO₂ in the atmosphere is shown figure 4.
The rate of formation of CO₂ and the rate of reduction of CO₂ in the atmosphere are not proportional. In time reduction of CO₂ is very much affected due to deforestation and increased human activities and the formation of CO₂ increased abnormally due to industrial revolution. The formation of CO₂ increases exponentially whereas the reduction is not observed in the same rate (6). This leads to a growing gap between the two processes and the increase in the residual CO₂ concentration in the atmosphere. This demands the need for fixing the CO₂ concentration in the atmosphere to avoid the impact of CO₂ on human health, environment and societal welfare.

**PRINCIPLE OF CO₂ FIXATION**

Carbon fixation, as explained in figure 5, refers to the process by which gaseous carbon dioxide is captured, extracted, utilized and fixed.
It refers mostly to the processes found in autotrophs (organisms that produce their own food), usually driven by photosynthesis, whereby carbon dioxide is changed into sugars. Carbon fixation can also be carried out by the process of calcification in marine calcifying organisms and also by heterotrophic organisms in some circumstances (7). Very limited CO₂ fixation also occurs in nature using light and other sources such as by algae and enzymes by converting into organic carbon compounds and biomass.

**NATURAL FIXATION**

**NATURAL OCEAN CARBON CYCLE**

The oceans contain about 50 times more CO₂ than the atmosphere and 19 times more than the land biosphere. CO₂ moves between the atmosphere and the ocean by molecular diffusion when there is a difference between CO₂ gas pressure (PₐCO₂) between the atmosphere and oceans. For example, when the atmospheric PₐCO₂ is higher than the surface of the ocean, CO₂ diffuses across the air-sea boundary into the sea water. The oceans are able to hold much more carbon than the atmosphere because most of the CO₂ that diffuses into the oceans reacts with the water to form carbonic acid and its dissociation products, bicarbonate and carbonate ions. The conversion of CO₂ gas into nongaseous forms such as carbonic acid and bicarbonate and carbonate ions effectively reduces the CO₂ gas pressure in the water, thereby allowing more diffusion from the atmosphere. The oceans are mixed much more slowly than the atmosphere, so there are large horizontal and vertical changes in CO₂ concentration. In general, tropical waters release CO₂ to the atmosphere, whereas high-latitude oceans take up CO₂ from the atmosphere. CO₂ is also about 10 percent higher in the deep ocean than at the surface.

**PHOTOSYNTHESIS**
Photosynthesis is the process of producing organic molecules using the energy of light. The reaction for the production of glucose, the most abundant product and equation is given below:

$$6\text{CO}_2 + 12\text{H}_2\text{O} \rightarrow C_6\text{H}_{12}\text{O}_6 + 6\text{H}_2\text{O} + 6\text{O}_2$$

A description of the photosynthesis process is illustrated in figure 6.

![Photosynthesis overview](image)

Figure 6: Photosynthesis overview

Factors limiting photosynthetic rates include light intensity, water availability, soil nutrient content, concentration of carbon dioxide and temperature. The light dependent pathways produce ATP and NADPH+H⁺ to be used in the light independent processes. The compound requiring the NADPH+H⁺ and ATP was found to be Glycerate 3-phosphate (GP). GP is converted to various other three carbon sugars or Triose phosphates (TP), some are used to produce hexodes such as fructose phosphate and glucose phosphate.

**ARTIFICIAL FIXATION**

**FIXATION BY ABSORPTION**

In principle, CO₂ is reduced from the gas by absorbing it into source and regenerating the source into its original. A typical example of the CO₂ reduction using absorption method from a combustion source is illustrated in figure 7.
Exhaust gas is emitted as a result of the combustion of fuels from the engine. The conventional catalytic converter promotes the conversion of hydrocarbon (HC), nitrous oxides (NOx) and carbon monoxide (CO) in the engine’s exhaust into carbon dioxide (CO$_2$), nitrogen (N$_2$) and water (H$_2$O) vapor (8). The CO$_2$ absorber will absorb the CO$_2$ and then it will dispatch into CO$_2$ free exhaust and the CO$_2$ absorber is regenerated. Different types of sorbents are used depending upon the source and the need.

**AMINES AS SORBENTS**

The simple method is to pass the gas through a chemical solvent that selectively absorbs only the CO$_2$ and keeps it in a weak chemical bond. The CO$_2$ releases from the chemical solvent during regeneration by heat addition that breaks the bond of the CO$_2$. The almost pure CO$_2$ is collected, compressed and sent to storage (9). The retrieved solvent goes for re-use. The most common chemical solvents used are amines. Amine capture is a proven system in the natural gas cleaning process. The scale of operations is much larger for the removal of CO$_2$ from power plants. Even though many pilot plants are in service, a commercially viable operation on a large scale is yet to take place.

**LITHIUM HYDROXIDE AS SORBENT**

Lithium Hydroxide (LiOH) is the most commonly used CO$_2$ sorbent for use in expendable devices. The presence of water vapor is important to the functioning of LiOH beds (10). Chemisorption of CO$_2$ is thought to take place via a two step reaction in which lithium
monohydrate is first formed by the exothermic reaction followed by the endothermic formation of lithium carbonate.

\[
\text{LiOH(s) + H}_2\text{O (g) } \rightarrow \text{ LiOH.H}_2\text{O(s)} \quad \Delta H^o = -14.5 \text{ kcal/mol LiOH}
\]

\[
2 \text{ LiOH.H}_2\text{O(s) + CO}_2\text{(g) } \rightarrow \text{ Li}_2\text{CO}_3\text{(s) + 3 H}_2\text{O (g)} \quad \Delta H^o = +3.8 \text{ kcal/mol LiOH},
\]

Significantly, water is required on both sides of these reaction equations. For the net reaction,

\[
2 \text{ LiOH + CO}_2\text{(g) } \rightarrow \text{ Li}_2\text{CO}_3\text{(s) + H}_2\text{O(s)} \quad \Delta H^o = -10.7 \text{ kcal/mol LiOH}
\]

two moles of water are liberated for each mole of CO\text{\textsubscript{2}} chemisorbed.

**CO\text{\textsubscript{2}} Fixation Using Catalyst**

Conventional catalysts such as activated alumina, magnesium oxide, graphene, natural zeolites, clays and zinc oxide are used to reduce CO\text{\textsubscript{2}}. The efficiency of these compounds is not enough to reduce the large quantity of CO\text{\textsubscript{2}} in the atmosphere. The turn over number, the number of molecules reacted per site per time is too low using these materials due to lower surface area and low activity.

More effective catalysts with larger surface area of efficient active sites need to be developed. Nanocatalysts are one such material providing higher activities. i.e., its incorporation in organic molecules such as formic acid requires a catalyst. Bulk CdS and ZnS surfaces catalyze CO\text{\textsubscript{2}} fixation in the presence of light, but CdSe surfaces do not. However, Cd-rich CdSe nanocrystal below a certain critical size is an efficient photo-catalyst (11). We report first-principle calculations that reveal distinct roles played by several different aspects of the nanoscale. On flat stoichiometric CdSe surfaces a CO\text{\textsubscript{2}} molecule physisorbs and is no more reactive than in the freestate. At a Cd vacancy, however, strong chemisortions occur, and the molecule draws extra electron density from the back bonds to become negatively charged. The barrier for desorption is ~ 0.3 eV suggesting that, even at room temperature, CO\text{\textsubscript{2}} molecules would be constantly chemisorbing and desorbing.
If a chemisorbed molecule could desorb and carry an extra electron with it, it would be highly reactive. Photo excitation, which excites electrons to the conduction bands, is essential for the catalytic process to occur. Doping the crystal n-type in the calculation reduces the energy cost to only 0.4 eV it is now that a nanocrystal enters the scene as an absolute necessity. The energy gap of a nanocrystal increases with decreasing size. The critical diameter to enable the free flow of crystal electrons to desorbing CO$_2$ molecules is estimated at about 3.5 nm, which compares well with the experimental value of 5 nm.

**CAPTURING BY ENZYME**

CO$_2$ solution based bioreactor contains an enzyme-bonded packing material that interacts with a water solution pumped in from the top and smokestack emissions that enter and bubble up through the bottom figure 8. The carbon dioxide in the emissions stream is captured by the enzymes on the surface of the packing material and converted into bicarbonate ions. Cleaned-up air then escapes from the top while the bicarbonate solution exits the bottom. The bicarbonate, in a separate process, can be extracted from the solution and made into compounds, such as limestone, for use by industry. A new way to capture carbon dioxide from smokestacks produces a raw material that can be sequestered underground or turned into substances such as baking soda, chalk, or limestone. The bioreactor is a long cylinder containing a packing material that acts as a solid support for the enzyme. The surface of this material has been chemically modified so that the enzymes attach securely. At the top of the cylinder, a water solution is pumped in and flows around the packing material, while gases from a smokestack enter the bottom of the cylinder and bubble up through the solution. The carbon dioxide is absorbed into the solution and then interacts with the enzymes, which convert the greenhouse gas into bicarbonate ions. To end the process, cleaned up air escapes from the top while the bicarbonate

<table>
<thead>
<tr>
<th>S.NO</th>
<th>Compounds</th>
<th>Surface Area (m$^2$/g)</th>
<th>Adsorption Rate (mmole/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>MgO</td>
<td>35 - 121</td>
<td>3.3 - 6.6 @ (20 torr)</td>
</tr>
<tr>
<td>2</td>
<td>Zinc Oxide</td>
<td>15</td>
<td>0.3125 @ (20 torr)</td>
</tr>
<tr>
<td>3</td>
<td>Aluminia Whiskers</td>
<td>320</td>
<td>0.625 @ (20 torr)</td>
</tr>
<tr>
<td>4</td>
<td>HEG</td>
<td>442.87</td>
<td>15 @ (11 bar &amp; 100°C)</td>
</tr>
<tr>
<td>5</td>
<td>PANI-t-HEG</td>
<td></td>
<td>31 @ (11 bar &amp; 100°C)</td>
</tr>
<tr>
<td>6</td>
<td>NAY</td>
<td>523 - 655</td>
<td>1.25 @ (20 torr)</td>
</tr>
<tr>
<td>7</td>
<td>BAY</td>
<td>332</td>
<td>1.23 @ (20 torr)</td>
</tr>
<tr>
<td>8</td>
<td>10 nm Alumina</td>
<td>195</td>
<td>0.3125 @ (20 torr)</td>
</tr>
<tr>
<td>9</td>
<td>Dry Water</td>
<td>0.813</td>
<td>3 times of water</td>
</tr>
</tbody>
</table>

Table 2: Catalysts used for CO$_2$ reduction
solution is extracted for further processing—either back into pure carbon dioxide for long-term geological storage or into a carbonate compound, such as limestone, that can be used by industry.

**Figure 8:** Capturing by enzyme

**FIXATION USING MICROALGAE**

Microalgae have attracted a great deal of attention for CO₂ fixation and biofuel production because they can convert CO₂ into biomass via photosynthesis at much higher rates than conventional bio-fuel crops can. Microalgae-mediated CO₂ fixation and biofuel production can be rendered more sustainable by coupling microalgae biomass production with existing power generation and wastewater treatment infrastructures figure 9.
Microalgae can utilize low-quality water, such as agricultural runoff or municipal, industrial or agricultural wastewaters, as a source of water for the growth medium as well as a source of nitrogen, phosphorus and minor nutrients.

Although the most common application of microalgae in wastewater treatment aims at nutrient removal/recovery, microalgae have also been utilized for removal of heavy metals and organic matter. Finally, secondary utilization of microalgae has been successful in toxicity monitoring. The combination of the three roles of microalgae CO$_2$ fixation, wastewater treatment and biofuel production has the potential to maximize the impact of microalgae biofuel production systems, and has accordingly been investigated (12 and 13). However, a number of crucial research gaps remain that must be overcome to achieve full-scale operation including: (i) improved algal growth and nutrient uptake rates; (ii) integration of biosystems with waste gas, wastewater and water reclamation systems; (iii) improved gas transfer and mixing; (iv) improved algal harvesting and dewatering; and (v) life cycle analysis (LCA) and associated economic assessment. In addition, there is a lack of fundamental information needed to rationally optimize the performance of existing bioreactors. Novel bioreactor configurations and designs are also needed that promote microalgae growth, characterized by volumetric productivities at least one order of magnitude above those of conventional open pond facilities.

**SEVERAL PATHWAYS EXIST FOR CONVERTING THE CO$_2$ INTO USEFUL PRODUCT**

Efficient conversion of carbon dioxide into useful materials continues to be a lingering energetic challenge. Researchers have attempted to convert carbon dioxide using a number of techniques which include but are not limited to:
1. Thermally induced conversion to mineral carbonates
2. Conversion of CO\textsubscript{2} to O\textsubscript{2} using algae ponds
3. Catalytic conversion to carbon dioxide into methanol, methane etc.

CONCLUSIONS

The reduction of carbon dioxide level in the atmosphere is an immediate need as CO\textsubscript{2} plays a significant role in the enhanced green house effect leading to abnormal climate change. The production of CO\textsubscript{2} is increased due to heavy use of fossil fuel and industrialization. The reduction of CO\textsubscript{2} is significantly reduced due to deforestation. The gap between the production and consumption of CO\textsubscript{2} concentration in the atmosphere is increased in time. Natural fixation of CO\textsubscript{2} in the atmosphere such as photosynthesis and ocean carbon cycle are enough to control the growing amount of CO\textsubscript{2}. Artificial fixation of CO\textsubscript{2} is necessary. Carbon dioxide is captured and extracted by various methods. Techniques using effective absorber, conventional and nano catalytic methods, capturing by enzymes and capturing using microalgae are few pathways to reduce CO\textsubscript{2}. The turn over number of CO\textsubscript{2} capture in the catalytic process is directly proportional to the surface area of the catalyst material and the nature of the active site of the catalyst. CO\textsubscript{2} is converted into methane, methanol, oxygen and bio mass using catalytic process. The continued challenge of converting CO\textsubscript{2} into useful material and the need for the large CO\textsubscript{2} reduction using a little amount of reducing material is a growing research. Taking into account the result of this study, we can see a significant influence of carbon dioxide fixation.

REFERENCES


AUTHORS

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Dr. S. Rajadurai is a corporate executive in the United States and India for over two decades. He has a mix of academic and industrial experience for the last 35 years. He received his Ph.D. (1979) in Physical Chemistry (Heterogeneous Catalysis) from Indian Institute of Technology, Chennai. He worked as an Assistant Professor of Chemistry at The American College, Madurai and Loyola College, Madras (1979-85). Dr. Rajadurai held research faculty position at the University of Notre Dame (1985-90). Dr. Rajadurai worked as a Research Leader and Director of Research at Cummins Engine Company and Molecular Technology Corporation (1990-96), Director of Advanced Development at Tenneco Automotive (1996-02) and Director of Emissions Systems at ArvinMeritor Inc. (2002-2004), Vice President of ACS Industries (2004-2009) and at Sharda Motor Industries since 2009. He is the author of more than 100 scientific papers, 130 Technical Reports and several patents on solid oxide solutions, free radicals, structure sensitivity of catalysts, mixed oxide catalysts, SCR and SNCR NOx reduction systems, and catalytic converter designs.

Dr. Rajadurai is a Fellow of the Society of Automotive Engineers. He is a life member of North American Catalysis Society, North American Photo Chemical Society, Catalysis Society of India, Instrumental Society of India, Bangladesh Chemical Society and Indian Chemical Society. He was the UNESCO representative of India on low-cost analytical studies (1983-85). He was awarded the Tenneco Innovation Award in 1998, 1999 for developing computer-aided tools for converter design and for validating low noble metal catalytic converter. He received the General Manager’s Leadership Award (1998) and also the 2000 Vision Award for developing strategies for cleaner, quieter, and safer transportation. Dr. Rajadurai is an active participant in Clean and Green Earth Day demonstrations since 1997 and US Clean Diesel School Bus Summit (2003) at Washington D.C. Dr. Rajadurai is a panelist of the Automotive R&D Scientists and Technologists of Indian Origin, New Delhi 2004.
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