

Though this document is called a whitepaper, this document is more of a letter to a prospective stakeholder talking about what we do, where we are and where we want to be. I accept that this document is not perfect, please accept my apologies. Please do not compare this document with a peer reviewed journal publication as this paper is quite informal. You will find instances where there are numbers and other information, but you may not find the reference - they are not absurd – if you search, you will find them. Also, I extend my pre-emptive apologies for any potential imprecision in my language usage, sometimes incoherent, or any incomplete information, and the utilization of imperfect analogical representations. If you identify any errors or instances of inaccurate information within the document, I welcome and appreciate your inputs for correction – Arnab Sinha

You can always write to me at arnab.sinha@novonanmek.com

(Picture Credit: royalsociety.org)

Legal Disclaimer

The information contained in this whitepaper is provided for general informational purposes only and does not constitute professional advice. The author makes no representations or warranties of any kind, express or implied, about the completeness, accuracy, reliability, suitability, or availability of the information contained herein. Any reliance you place on such information is strictly at your own risk.

In no event will the author be liable for any loss or damage including, without limitation, indirect or consequential loss or damage, or any loss or damage whatsoever arising from loss of data or profits arising out of, or in connection with, the use of this whitepaper.

Through this whitepaper, you are able to link to other websites that are not under the control of the author. The author has no control over the nature, content, and availability of those sites. The inclusion of any links does not necessarily imply a recommendation or endorse the views expressed within them.

Every effort is made to keep the whitepaper up and running smoothly. However, the author takes no responsibility for, and will not be liable for, the whitepaper being temporarily unavailable due to technical issues beyond our control.

Arnab Sinha

Novonanmek Material Sciences Private Limited

Objective of this Whitepaper

The primary objective of this whitepaper is to provide a comprehensive overview of our innovative approach to carbon capture using pelletized calcium oxide (CaO) enriched with additives. Through this whitepaper, we aim to share our research findings, insights, and advancements in the field of Direct Air Capture (DAC) technology. Our focus is on leveraging the synergistic effects of carefully selected additives to enhance the reactivity and efficiency of CaO pellets for capturing CO2 from ambient air. By presenting the methodology, benefits, and potential applications of this technology, we seek to contribute to the discourse surrounding sustainable carbon capture solutions. We also provide an overview of how the we utilize the captured CO2 and, in the process, decarbonize Lithium mining and refining operation.

Our goal is to engage with researchers, practitioners, industry stakeholders, and policymakers interested in advancing the development of environmentally responsible and economically viable carbon capture methods. We believe that by sharing our knowledge and innovation, we can collectively work towards addressing the challenges of climate change and fostering a greener future.

Table of Contents:

1.	Summary	5
2.	Introduction	5
3.	Technology Overview	6
4.	Laboratory work	8
5.	Process Description	15
6.	Pilot Plant	18
7.	Scaling Up	21
8.	Utilization of captured CO ₂	22
9.	Economics	24
10.	Conclusion	25
11.	Future Work	25

1. Summary

This document unveils a novel approach to Direct-Air-Capture (DAC) based on the Calcium Looping (CaL) cycle. The crux of this innovative technology lies in the careful selection of additives, which function as catalysts, promoters, or modifiers. These additives serve to significantly enhance the interaction between CO2 and CaO, constituting a pivotal aspect of the process. Incorporation of these chosen additives occurs during the formation of pellets within the CaO matrix.

The pelletization procedure ensures a uniform dispersion of additives throughout the CaO structure. This results in a harmonious amalgamation that optimizes reactivity, thereby substantially bolstering carbon capture efficiency.

Furthermore, the document introduces a method for effectively utilizing the captured carbon dioxide. In this method, the CO2 captured during the process is introduced into an aqueous solution of Lithium Hydroxide (LiOH). This interaction facilitates the conversion of Lithium Hydroxide into Lithium Carbonate (Li2CO3). Remarkably, this process contributes to the prevention of approximately 10.8 tonnes of CO2 from being released into the atmosphere.

Moreover, the procedure unveils another remarkable potential: when half of the LiOH output from a refinery is allocated for conversion into Lithium Carbonate through our method, it becomes feasible to achieve full decarbonization of the entire mining and refinery operation. This achievement holds particular significance, given the emissions associated with both hard rock lithium resources and refining facilities.

Furthermore, the revelation emerges that this process yields a substantial gross margin. This margin is substantial enough to completely counterbalance the expenses of Direct-Air-Capture per metric ton of removed Carbon Dioxide, all while maintaining profitability.

2. Introduction

The global challenge of mitigating climate change has intensified the search for innovative and efficient methods of capturing carbon dioxide (CO2) from the atmosphere. Direct Air Capture (DAC) has emerged as a promising solution, offering the potential to remove CO2 directly from the air and contribute to the reduction of greenhouse gas emissions. Within the realm of DAC, our focus lies on a groundbreaking technology that leverages the power of pelletization, utilizing calcium oxide (CaO) as a solid sorbent, enhanced with carefully selected additives.

As the urgency to address climate change heightens, the need for scalable and efficient carbon capture technologies becomes more evident. This white paper delves into a novel approach that not only harnesses the inherent reactivity of CaO but also capitalizes on the synergistic effects of carefully chosen additives. By pelletizing CaO with precision-engineered additives, we aim to accelerate the carbonation process at ambient temperature, unlocking a cost-effective and energy-efficient pathway to capture CO2 from the air.

In this comprehensive white paper, we will explore the intricacies of this technology, from its fundamental principles to its potential applications in diverse sectors, including power, steel, cement plants, and beyond. We will delve into the additive selection process, pellet formulation, optimization strategies, and the results of experimental studies that validate the effectiveness of this approach. Additionally, we will discuss the advantages, challenges, and

opportunities associated with this pioneering technique, shedding light on its scalability, economic viability, and potential contributions to a sustainable future.

Join us on this journey of innovation and discovery as we unveil a path forward in the realm of carbon capture. Through the fusion of science, engineering, and environmental stewardship, we aspire to reshape the landscape of carbon capture technologies, contributing to the collective efforts to combat climate change.

3. Technology Overview: Enhanced Carbon Capture through Innovative Pelletization

This is a solid sorbent Direct-Air-Capture (DAC) technology based on the calcium looping (CaL) cycle.

Direct Air Capture (DAC) has emerged as a transformative approach in the battle against climate change, offering the potential to directly remove carbon dioxide (CO2) from the atmosphere. Central to the effectiveness of DAC is the selection of suitable sorbents that can efficiently capture and retain CO2. In this pursuit, the utilization of calcium oxide (CaO) has gained considerable attention due to its inherent reactivity with CO2. However, the challenge lies in enhancing the kinetics of the carbonation process at ambient temperature, a crucial factor for making DAC feasible on a large scale.

Our innovative approach centers on the pelletization of CaO with carefully engineered additives, aiming to not only boost the reactivity of CaO but also accelerate the carbonation process, all while occurring at ambient temperature. This technology represents a critical advancement in the field of DAC, offering a promising solution to the challenges posed by slow reaction rates and energy-intensive processes.

Key Elements of the Technology

- a. Additive Selection: The heart of our technology lies in the selection of additives that act as catalysts, promoters, or modifiers to enhance the interaction between CO2 and CaO. These additives are chosen based on their ability to facilitate CO2 adsorption and reaction, leading to faster carbonation rates.
- b. Pellet Formulation: The chosen additives are incorporated into the CaO matrix during pellet formation. The pelletization process ensures a homogenous distribution of additives within the CaO structure, creating a synergistic blend that optimizes reactivity.
- c. Optimization Strategies: Pellet formulation parameters, such as additive concentration, particle size, and mixing techniques, are meticulously optimized to achieve the desired balance between reactivity enhancement and mechanical integrity of the pellets.
- d. Enhanced Carbonation at Ambient Temperature: Thanks to the additives' inclusion along with controlled moisture, the resulting pellets exhibit a remarkable ability to rapidly undergo carbonation at ambient temperature. This means that CO2 capture occurs without the need for energy-intensive high-temperature processes.

Advantages of the Technology:

- a. Enhanced Reactivity: The addition of additives accelerates the carbonation process, enabling significant CO2 capture at lower temperatures.
- b. Energy Efficiency: The technology operates at ambient temperature, reducing energy consumption compared to conventional high-temperature methods.
- c. Scalability: The pelletized CaO approach offers scalability, with potential applications ranging from small-scale DAC units to large industrial installations.
- d. Versatility: The technology can be applied to various sectors, including DAC, power, steel, and cement plants, offering a versatile solution for diverse emission sources.

We did not directly arrive at this technology, but after almost four years of experimentation with various processes like bubbling atmospheric air thorough aqueous solution of alkalis (not only sodium or potassium hydroxides, but also some high-demand materials like Lithium Hydroxide – we have used the process with LiOH in utilizing the captured CO2), bubbling atmospheric air thorough UV illuminated aqueous dispersion of photocatalytic nanomaterials, bubbling atmospheric air though sea water with increased alkalinity - all the processes in carefully designed high throughput bubble reactors with base plates having multiple orifices for pushing air into the reactor containing the above mentioned liquids at a very high rate, and other process using solid sorbents, we found this technology to be easily adaptable and scalable. With other technologies, I could not see beyond a certain point in the future, but this is the only technology, with which I am able to see at least 200 years into the future. This is obvious for one reason – **availability of Limestone**.

This technology is standing on two major things – Limestone and Energy. Limestone is available in abundance and energy can always be generated from renewable sources. There is no dependence on another company for products like specialty sorbents. *So, there is nothing that can hold this technology back.*

Calcium looping is a technology used for carbon capture and storage (CCS) in which carbon dioxide (CO2) is captured from flue gas or atmospheric air by using calcium oxide (CaO) as a sorbent. The process involves two main chemical reactions:

- 1. Carbonation Reaction: CaO + $CO_2 \rightarrow CaCO_3$
- 2. Calcination Reaction: $CaCO_3 \rightarrow CaO + CO_2$

In these equations:

CaO: Calcium oxide (also known as quicklime or lime) is the sorbent material.

CaCO₃: Calcium carbonate is the product of the carbonation reaction, and it is also known as limestone.

CO₂: Carbon dioxide is the greenhouse gas that is captured from the flue gas or atmospheric air in case of Direct-Air-Capture (DAC)

During the carbonation reaction, calcium oxide reacts with carbon dioxide to form calcium carbonate. This is the step where CO2 is captured from the flue gas. During the calcination

reaction, calcium carbonate is heated to release the captured CO2 and regenerate the calcium oxide for reuse.

The cyclic process involves alternating between these two reactions to capture and release CO2. This technology has potential applications in reducing CO2 emissions from industries such as power generation, cement production and now Direct-Air-Capture (DAC).

4. Laboratory Work

Limestone lumps (about 10 Kgs) were heated in an electrical furnace to a temperature little over 1100°C and held the material at 1100°C till all the CO2 was released from CaCO3 and CaO was left behind. The CO2 that was released was not captured the first time.

The CaO that was obtained was mostly in powder form with some colour change. Powders cannot be exposed to even air flowing in a laboratory oven, let alone fast flowing air being blown by a high-power fan that would suck in atmospheric air in Direct-Air-Capture systems. So, we decided to press the powder into square blocks using a manual laboratory press. The CaO powder was mixed with binders and using a mould was pressed into square blocks of size approximately 1 Inch x 1 Inch x 1 Inch. (each weighing about 49 grams).

These blocks were weighed, their individual weights noted and placed in a laboratory oven at a temperature of 70°C with air being blown over the samples for over 10 hours. Intermittently the sample weights were noted to see if any weight gain and reach a point where there is no further weight gain. It is to be noted that as atmospheric air containing CO2 was blown over the samples, the CO2 would react with CaO samples and form CaCO3 – this is what our objective is. We would want all the CaO to react with atmospheric CO2 in minimum time. But is it so easy? We repeated the experiments several times by increasing and decreasing the moisture level in the pressed samples and also by increasing and decreasing the air supply through the blower, which is a part of the oven and used to blow air inside a laboratory oven through the samples.

The following were the observations:

- 1. Morphology of CaCO3 powder change during calcination as it released CO2 and turned into CaO Lumps change into powder. There was also some change in the colour.
- 2. The Reaction of CaO and CO2 can take place at ambient temperature in the presence of moisture.
- 3. Presence of Moisture enhances the CaO CO2 reaction
- 4. Pressing powder CaO into pellets can withstand high velocity air that can accelerate the CO2 capture
- 5. An optimal Packing factor will allow smooth passage of fast flowing air through the pellets.
- 6. The pellets are observed to gain weight will being pelletized but this is very minor. This is indicative of the fact that a very small conversion of CaO takes place even during the pelletization process.

- 7. Pellets might crack or lose integrity when subjected to high-speed air flow or during sudden change in temperature.
- 8. A layer of CaCO3 forms on the CaO pellets during carbonation process that could prevent further reaction of CaO with CO2.

Based on the above observations, we concluded the following:

- 1. High reaction kinetics will lead to maximum CaO conversion to CaCO3 this will ensure proper carbon capture.
- 2. After calcination of CaCO3, the powdered CaO cannot be directly subjected to high velocity air, so we need to **turn the powder into pellets** *this and additive selection are the two most important step in our Direct-Air-Capture (DAC) process.*

By using pelletized CaO in Calcium Looping (CaL) for Direct Air Capture (DAC), we are addressing several challenges and improving the efficiency of the process:

- a. Enhanced Carbon Capture Efficiency: Pelletized CaO offers a higher surface area compared to traditional bulk CaO. This increased surface area facilitates more effective contact between the CaO pellets and the surrounding air, enhancing the rate of CO2 capture during the carbonation phase of the CaL process.
- b. **Improved Reactivity**: Pelletization can lead to better control over the porosity and particle size distribution of the CaO pellets. This optimization can enhance the reactivity of CaO with CO2, resulting in higher CO2 capture rates and requiring less energy for carbonation.
- c. **Reduced Energy Consumption**: The improved reactivity and enhanced surface area of pelletized CaO can potentially lead to a reduction in the energy required for the calcination and carbonation steps. This can make the overall CaL process more energy-efficient and economically viable.
- d. **Controlled Particle Size:** Pelletization allows for precise control over the size and shape of the CaO pellets. This can lead to consistent performance and improved handling during the process.
- e. **Minimized CO2 Absorption Limitations:** In DAC, the availability of CO2 in the atmosphere is limited due to its low concentration. The improved efficiency and reactivity of pelletized CaO can help overcome this limitation by maximizing the CO2 capture from the limited amount of air passing through the pellets.
- f. **Potential Scalability:** Efficient and optimized pelletized CaO can contribute to the scalability of DAC technologies by improving the overall process efficiency and reducing the energy consumption per unit of CO2 captured.
- We must make sure that the reaction between CaO and CO2 takes place at a temperature that is little higher than ambient temperature – this will require selection of the right additive for the CaO pellets.
- 4. The right geometry of the pellet and the right packing factor will allow smooth and fast passage of fast flowing air through the pellets and thus increasing the CO2 capture

- 5. The pellets must be strong enough to withstand the force of high velocity air this will require selection of the right additive for the CaO pellets.
- 6. We must make sure that even though CaCO3 forms on the surface, the reaction between CaO and CO2 continue to take place in the interior of the pellets. - this will again require selection of the right additive for the CaO pellets.

To achieve the above, we decided to turn pure CaO pellets into composite pellets with the base material as CaO and other additives distributed evenly in the base matrix.

The following additives were studied before we could finally arrive at a composition that offers all the necessary characteristics of the pellets. This is the one of the two most important parts of this project. The other one is pelletization.

- 1. Alkaline Earth Metal Oxides (E.g.: MgO): These are promoters. They facilitate faster or more complete carbonation of CaO.
 - a. Enhanced Reactivity: Alkaline earth metal oxides, like MgO, can exhibit higher reactivity with CO2 compared to pure calcium oxide (CaO). When mixed with CaO, these oxides can create a composite material that has improved CO2 capture kinetics. This means that the carbonation reaction—the conversion of CaO to CaCO₃—can occur more rapidly, enhancing the efficiency of the process.
 - b. Lower Carbonation Temperature: The presence of alkaline earth metal oxides can lower the temperature at which CO2 capture takes place. This is particularly advantageous if you're aiming to capture CO2 at lower temperatures, as it can reduce the energy requirements and potentially allow for more flexible process conditions.
 - c. Cyclic Stability: Composite materials containing alkaline earth metal oxides may exhibit improved cyclic stability. This means that the material can undergo repeated cycles of carbonation and calcination without significant degradation in its CO2 capture capacity. This durability is essential for the long-term effectiveness of carbon capture processes.
 - d. Synergistic Effects: The interaction between calcium oxide and alkaline earth metal oxides can create synergistic effects that enhance the overall carbon capture performance. These interactions can lead to more accessible active sites for CO2 adsorption and more efficient utilization of the material.
 - e. Process Flexibility: The addition of alkaline earth metal oxides can offer more flexibility in process conditions, including temperature and CO2 concentration, without compromising carbon capture efficiency.
 - f. Improved Pellet Properties: When using these oxides during pelletization, they can contribute to the mechanical strength and structural integrity of the pellets, further enhancing their performance during the carbonation step.

2. Alkali Silicates (E.g.: Sodium silicate (Na2SiO3) and potassium silicate (K2SiO3)

These are binders that enhance carbon capture.

Alkali silicates are compounds that contain alkali metals (such as sodium, potassium, or lithium) and silicate ions. When added to CaO pellets for carbon capture, alkali silicates can play a role in enhancing the performance of the pellets by improving reactivity, pore structure, and resistance to sintering. These compounds can facilitate CO2 absorption and release during carbonation and calcination cycles. While there are various types of alkali silicates, sodium and potassium silicates are often considered for carbon capture applications.

- a. Reactivity Enhancement: Alkali silicates can promote the reactivity of CaO with CO2, leading to more efficient carbonation and calcination processes.
- b. Pore Formation: Alkali silicates can contribute to the formation of a porous structure within the pellets, enhancing the surface area available for CO2 absorption and release.
- c. Resistance to Sintering: Alkali silicates can help prevent excessive sintering of pellets during calcination, maintaining pellet integrity and preventing particle agglomeration.
- d. Binding and Cohesion: Alkali silicates can act as binders that help maintain the structural integrity of pellets during handling, calcination, and carbonation cycles.
- e. Controlled Release: Alkali silicates can facilitate controlled release of CO2 during carbonation and efficient reabsorption during calcination, optimizing the capture and release processes.

3. Organic and Physical Pore Formers (E.g., Starch, Polystyrene beads)

- **a.** Pore Forming Agents These can enhance the porosity of the pellets, increasing surface area and improving CO₂ diffusion into the pellet
- **b.** Porosity Enhances Surface Area, leads to improved gas diffusion, and increased CO2 absorption.

4. Nanostructured Materials:

Nanoparticles: Nanoparticles of materials like silica (SiO2), alumina (Al2O3), or titania (TiO2) can be incorporated into the pellet matrix.

Nanofibers: Nanofibers made from materials like carbon nanotubes (CNTs) or cellulose can be mixed into the pellet composition.

Nanoporous Materials: Zeolites and other Nanoporous materials can be included to introduce additional porosity to the pellet structure

Nanoparticle Aggregates: Aggregates of nanoparticles, such as colloidal silica aggregates

These nanostructured surface area enhancers work by providing more active sites for gas adsorption, allowing gases to diffuse into the internal porosity of the pellets, and

creating a more complex and intricate surface structure. The choice of enhancer depends on factors such as compatibility with the pellet matrix, reactivity with CO2, and overall pellet performance.

These are Surface Area Enhancers and they Increase surface area can lead to more contact points for CO_2 , potentially speeding up carbonation.

- a. These nanoparticles have a high surface area-to-volume ratio due to their small size. When distributed within the pellet, they create additional sites for CO2 adsorption and reaction.
- b. Nanofibers create a three-dimensional network that increases the pellet's surface area and porosity. They can also enhance mechanical strength.
- c. Nanoporous materials have intricate networks of nanoscale pores, which can act as reservoirs for CO2 capture.
- d. Nanoparticle Aggregates can be used to introduce micro- and nanoscale roughness to the pellet surface, enhancing gas-solid interactions.

5. Hydroxides (E.g.: Sodium & Potassium Hydroxide)

- a. Materials that can react with CO₂ to form carbonates can be mixed with CaO to provide dual absorption mechanisms. Aqueous solution of NaOH shall be used to spray over the CaO pellets during carbonation in the reactor.
- 6. Clays, Alumina, Zirconia: Increase mechanical strength and integrity
- 7. **Zirconia & Cerium Oxide:** Thermal stabilizers, help prevent particle sintering and agglomeration during carbonation & Calcination

Several other materials that were studied can be included in pellets to enhance CO2 capture efficiency. These materials can serve various purposes, such as increasing surface area, improving reactivity, and providing stability. Some of them are below:

- a. **Silica (SiO2)**: Silica is known for its high surface area and porosity. Adding silica nanoparticles or a silica-based material can increase the overall surface area of the pellets, enhancing CO2 adsorption capacity.
- b. **Clays**: Clays like montmorillonite and kaolinite can offer improved CO2 capture efficiency due to their layered structure, which provides more sites for CO2 adsorption.
- c. **Activated Carbon**: Activated carbon has a highly porous structure that can significantly increase the available surface area for CO2 adsorption.
- d. **Zeolites**: Zeolites are microporous materials that can selectively adsorb CO2. Adding zeolites to the pellets can enhance the capture of CO2 from gas streams.
- e. **Metal-Organic Frameworks (MOFs**): MOFs are highly porous materials that can be designed with specific properties for CO2 capture. Incorporating MOFs into pellets can boost adsorption efficiency.

- f. **Aluminium Oxide (Al2O3):** Aluminium oxide can enhance the stability of the pellets and potentially contribute to CO2 capture efficiency.
- g. **Fly Ash:** Fly ash, a byproduct of coal combustion, can be utilized as an additive. It contains oxides that can react with CO2, contributing to carbonation.
- h. **Nanostructured Materials:** Various nanostructured materials, such as nanoparticles or nanofibers, can be incorporated to increase reactivity and surface area.
- i. **Waste Byproducts:** Certain waste materials, when processed and incorporated, can contribute to CO2 capture while also addressing waste management concerns.
- j. **Catalysts:** Catalysts can accelerate the carbonation reaction, improving the efficiency of CO2 capture.

Below is a list of catalysts that were also explored before we could zero down on our final composition.

- a. **Transition Metal Oxides**: Metal oxides like iron oxide (Fe2O3), nickel oxide (NiO), and cobalt oxide (CoO) can catalyse the carbonation reaction, enhancing the conversion of CaO to CaCO3.
- b. Alkali Metal Compounds: Alkali metal compounds, such as potassium carbonate (K2CO3) or sodium hydroxide (NaOH), can improve reaction rates and increase the efficiency of CO2 capture.
- c. Alkaline Earth Metal Oxides: In addition to MgO, other alkaline earth metal oxides like strontium oxide (SrO) and barium oxide (BaO) have also been considered as catalysts.
- d. **Metal-Organic Frameworks (MOFs):** Certain MOFs with specific metal centres can act as catalysts, enhancing CO2 adsorption and reaction kinetics.
- e. **Enzymes and Biomimetic Catalysts:** Biological catalysts or biomimetic catalysts inspired by enzymes have been explored for their potential to enhance CO2 capture processes.
- f. Nanostructured Catalyst: Nanostructured catalysts, such as nanoparticles or nanowires, can provide high surface area and reactivity, improving CO2 capture efficiency.
- g. **Promoters:** Some materials act as promoters to enhance the performance of catalysts. For example, promoting catalysts with certain alkali metals can boost their activity.
- h. **Mixed Metal Oxides:** Combining different metal oxides or compounds can lead to synergistic effects that improve catalytic activity.

There is another class of additive that was studied called **Micronutrient Additives**.

Micronutrient additives are materials that provide essential trace elements required for the growth and health of plants. While their primary role is to support plant growth, some micronutrients can also influence pellet properties, stability, and potentially CO2 capture performance. One commonly used micronutrient that could potentially be considered for inclusion in a CaO pellet composition is iron.

Iron (Fe): Iron is an essential micronutrient for plant growth and plays a crucial role in various biochemical processes. It's often included in fertilizers to prevent iron deficiency in plants. In pellet compositions, a small amount of iron-based compound (**such as iron oxide**) could be added. Iron can contribute to pellet stability and potentially interact with other components, although its influence on CO2 capture might be secondary compared to the main components like CaO and catalysts.

After rigorous experimentation with numerous compositions of the pellets, we finally arrived at our final composition of the Pellet for Direct-Air-Capture. Let us call this **DAC-PC1 (Direct-Air-Capture – Pellet Composition 1)**. This composition is our intellectual property or trade secret that cannot be disclosed in this paper We have also modified the pellet composition so that they can also be used to capture carbon dioxide from flue gases. But we are not concentrating on it right now. We will focus on Direct-Air-Capture (DAC).

Experimental Procedure: 10 Samples each of CaO and DAC-PC1 pressed into sizes of 1 Inch x 1 Inch x 1 Inch, soaked in 1 M solution of NaOH were kept on petri dishes and were kept in a hot air oven at a temperature of 70° C with air being blown over the samples. NaOH aqueous solution was added to both the samples manually with a dropper every 1 hour to keep the samples soaked in NaOH aqueous solution.

The experiment was conducted for 10 hours. After 10 hours, it was observed that both the samples no longer existed in their original form and shape but existed in the form of a precipitate in the solution of NaOH in the petri dish.

The solution was filtered to separate out the CaCO3, dried and weighed.

Following are the results that compare the carbon capture potential of CaO and CaO-Composite (DAC-PC1) pellets:

	Tests with pressed blocks of Pure CaO powder					
SL	Sample No.	Initial	Final	Weight in case of	Carbonation	
No.		Weigh	Weight (g)	complete carbonation	(%)	
		t (g)		of CaO (g)		
1	CaO-1	49.23	57.96	87.63	66.14	
2	CaO-2	50.09	56.95	89.16	63.87	
3	CaO-3	49.86	56.69	88.75	63.88	
4	CaO-4	49.12	58.83	87.43	67.29	
5	CaO-5	49.76	58.57	88.57	66.13	
6	CaO-6	49.82	56.64	88.68	63.87	
7	CaO-7	49.64	56.43	88.36	63.86	

Table 1: Laboratory Data showing extent of carbonation of CaO pellets

8	CaO-8	49.00	56.69	87.22	65.00
9	CaO-9	49.51	57.28	88.13	65.00
10	CaO-10	49.41	57.17	87.95	65.00

Table 2: Laboratory	Data showing	extent of	Carbonation	of DAC-PC1	pellets
---------------------	--------------	-----------	-------------	------------	---------

Tests with pressed blocks of DAC-PC1 powder					
SL	Sample No.	Initial	Final	Weight in case of	Carbonation
No.		Weigh	Weight (g)	complete carbonation	(%)
		t (g)		of CaO (g)	
1	DAC-PC1-1	52.11	81.92	84.63	96.80
2	DAC-PC1-2	52.47	82.48	85.21	96.79
3	DAC-PC1-3	52.41	80.38	85.11	94.44
4	DAC-PC1-4	52.52	82.55	85.29	96.78
5	DAC-PC1-5	52.85	83.07	85.83	96.79
6	DAC-PC1-6	52.17	80.01	84.72	94.44
7	DAC-PC1-7	52.28	83.18	84.90	97.97
8	DAC-PC1-8	52.75	83.91	85.67	97.95
9	DAC-PC1-9	51.97	82.70	84.40	97.99
10	DAC-PC1-10	51.87	81.54	84.24	96.80

(Please note that the powder DAC-PC1 has around 80% CaO and the rest are additives. During carbonation, only the CaO in the composite get carbonated to CaCO3, the rest get transformed differently).

The samples of DAC-PC1 have shown marked improvement in Carbonation Capacity than pure CaO samples. XRD studies have confirmed conversion of more than 96% of CaO to CaCO3 in DAC-PC1 samples.

5. Process Description

Our process is cyclic with 4 steps:



Fig 1: Our Four Step Cyclic Calcium Looping based Direct-Air-Capture (DAC)

- Calcination of Limestone: Limestone lumps are heated in an electrical furnace at a temperature of 900°C – 1100°C till the time no more CO2 is released. This is an energy intensive process, but if we have decided to use the Calcium Looping (CaL) Cycle, we have to bear with this and find other ways to reduce energy consumption or make CaO absorb sufficient carbon during the carbonation cycle that brings the energy consumption per tonne of Carbon Dioxide captured down.
- 2. Inclusion of Additives in the CaO Powder and Ball Milling: In this step we include some special materials into the CaO powder to make the composition that of DAC-PC1 and high energy ball mill the composite powder.

Ball milling is an important step in the process. The high energy ball milling process not only uniformly distributes the additives in the CaO matrix, but also the mechanical activation of CaO through high-energy ball milling creates defects and enhance the reactivity of CaO for carbonation. The inclusion of additives and the advantages it brings to the carbon capture process has been extensively discussed earlier in this document.

3. Pelletization: In this process, the composite powder containing CaO as the matrix is turned into pellets. As described before, the question of pelletization arises as CaCO3 turns into CaO powder post calcination and powder cannot be subjected to high velocity air as in the case of Direct-Air-Capture (DAC). This step of pelletization and inclusion of additives are the two most important steps in our process.

Pelletization offers several advantages when applied to the calcium looping (CaL) process for carbon capture. Here are some key benefits of using pelletized CaO in CaL:

- a. Enhanced Reactivity: Pelletization can lead to increased surface area and porosity of the CaO pellets, which enhances the reactivity between CaO and CO2. This means that the carbonation reaction can occur more efficiently and at a faster rate, improving overall capture performance.
- b. Controlled Geometry: Pelletization allows for precise control over the shape and size of the pellets. This enables better packing and stacking in reactor beds, ensuring optimal air flow and contact between the pellets and the gas stream.
- c. Mechanical Strength: Pelletization can improve the mechanical strength and durability of the CaO pellets. This is important for maintaining pellet integrity during handling, transport, and cycling between carbonation and calcination stages.
- d. Reduced Attrition: Pelletized CaO is less prone to attrition (breakage) compared to bulk CaO. This leads to reduced fines generation and loss of material, contributing to longer pellet lifespan and better process stability.
- e. Optimized Particle Size Distribution: Through pelletization, you can control the particle size distribution of the pellets. This can optimize the balance between surface area for reaction and gas flow resistance.

- f. Uniform Composition: Pelletization ensures a more uniform distribution of additives and binders within the pellet structure. This consistency leads to uniform reactivity across the pellet bed, reducing the risk of channelling and uneven CO2 capture.
- g. Tailored Additive Incorporation: Pelletization enables effective incorporation of additives that enhance CO2 capture performance, such as porosity-enhancing agents, surface area enhancers, and CO2 absorption promoters.
- h. Versatility: Pelletization can accommodate a wide range of CaO-based compositions and additives, making it suitable for tailoring pellets to specific process conditions and requirements.
- i. Easy Handling and Transport: Pelletized CaO is easier to handle, transport, and distribute within a reactor system compared to bulk materials. This simplifies operational logistics and reduces maintenance efforts.
- j. Process Integration: Pelletization can be seamlessly integrated into existing CaL processes and reactor designs, making it a viable retrofit option for improving carbon capture efficiency.

Overall, pelletization enhances the performance and efficiency of the CaL process for carbon capture by addressing key challenges associated with material reactivity, durability, and process stability.

4. Carbonation: Carbonation is the step when CaO Composite (containing additives) pellets are exposed to carbon dioxide in the atmospheric air and in the process gets converted to CaCO3. In our case carbonation happens in a special reactor which is vertical and can facilitate passing of high velocity atmospheric air through the reactor containing CaO-composite pellets. In our case, carbonation happens at around 40-70 °C in the presence of moisture. In the reactor the size, geometry, arrangement of pellets and the interstitial gaps play a major role as it makes the atmospheric CO2 come in close contact with the pellets in the reactor. The pellets have been arranged in such a way that the high-speed air touches almost every available surface while it flows through the reactor converting CaO to CaCO3 and in the process capturing CO2.



Reactor Design: Low Temperature Carbonation

6. Pilot Plant



Our pilot plant is planned to have a 600t per year capacity.

Fig 3: Schematic of our Calcium Looping (CaL) based Direct-Air-Capture Process

1. **Calcination:** 1100 ^oC Electrical Furnace having powered by Sandvik Kanthal APM Heating Elements having a capacity of 2-3 Tonnes.

During our Calcination Reaction (CaCO₃ \rightarrow CaO + CO₂), 2.27 Tonnes of Limestone (CaCO3) will he heated to release 1 tonne of CO2 and leave behind 1.27 Tonnes of CaO.

- High Energy Ball Mill: A high energy ball mill with capacity 3 tonnes would be necessary. Before or during the process of ball milling additives will be added to the CaO powder in the right proportion and ball milled for an even distribution of additives in the CaO matrix.
- 3. **Pelletization:** Our pellet geometry would be *hollow cylinders*. The size of the pellet with have a corelation with the size of the reactor where the pellets would be carbonated. The formula that connects the pellet size with the size of the reactor has not been disclosed in this paper as it is our trade secret.

Creating hollow cylindrical pellets would require specialized manufacturing process and equipment. Our pellets shall be made by extrusion. Here's how the process would work and the equipment involved:

- a. Material Preparation: Preparation of a mixture of the base material (CaO or CaO-based composite) along with any additives and binders. This mixture will be thoroughly mixed and homogenized.
- b. Extrusion: The prepared mixture will be fed into an extrusion machine, also known as an extruder. The extruder consists of a barrel with a rotating screw

inside. As the mixture enters the extruder, the rotating screw forces the material through a specially designed die at the end of the barrel.

- c. Die Design: The die used for hollow pellet extrusion will be designed to create the desired hollow cylindrical shape. The die includes an inner mandrel or core that shapes the hollow core of the pellet, while the surrounding space defines the outer shape of the pellet.
- d. Formation: As the material is forced through the die, it takes on the shape of the die's cavities, resulting in hollow cylindrical pellets. The hollow core is formed by the mandrel, and the material around it forms the pellet walls.
- e. Cutting and Sizing: The extruded material is then cut to the desired length using a cutting mechanism. The hollow cylindrical pellets are formed as individual units.
- f. Drying and Curing: The freshly extruded pellets may need to be dried and cured to remove excess moisture and allow any binders to set properly.

Equipment: Extrusion Machine (Extruder)

An extrusion machine (extruder) is the key equipment used in the hollow pellet extrusion process. The extruder consists of the following components:

- a. Barrel: A cylindrical chamber through which the material is fed and transported by the rotating screw.
- b. Screw: A rotating screw inside the barrel that pushes and compresses the material as it moves forward.
- c. Die: The die is located at the end of the barrel and shapes the material into the desired hollow cylindrical form.
- d. Cooling System: Depending on the material and process, cooling systems may be used to prevent overheating during extrusion.

The design and dimensions of the die will be critical to achieving the desired hollow cylindrical pellet shape. The extrusion process can be customized based on the specific requirements of the pellet composition and intended application.

4. Carbonation: During this process, high velocity air will be passed through a reactor containing the pellets (about 1.5t) made in the previous process, arranged in a way so as to allow fast moving air to pass though the interstitial gaps. pellets have been designed to allow fast flowing air and at the same time provide the surface or proper reaction of CaO with CO2 contained in atmospheric air leading to formation of CaCO3 and in the process capture of CO2.

The reactor will be around 4 m in diameter and around 2 m in height containing a 1.5t of CaO-composite pellets, fitted with four fans at the top sucking atmospheric air at a rate of 50,000 m³/hour each making air sucking capacity of 200,000 m³/hour, pushing air through the pellets in the reactor. During the process of carbonation, around 1.27t of CaO would react with 1 tonne of CO2 to form 2.27t of CaCO3, which will again undergo calcination. The pellets shall be sprinkled with water to moisten the pellets

and the temperature of the pellets raised to around 70 $^{\rm 0}{\rm C}$ to enhance the CaO-CO2 reaction.

- One tonne of CO2 = 1,000,000 g CO2
- Since the CO2 content in the air is 0.75g/m³ of air, 1,000,000g of CO2 will be contained in (1,000,000/0.75) m³ of air = 1.34 million m³ of air
- At a rate of 200,000 m3 per hour, it will take (1.34 million m³/200000 m³/hour) = 6.67 Hours to process 1 tonne of CO2.
- Assuming that only 50% of the CO2 passed though the reactor will be captured, it will take 13.3 hours to capture a tonne of CO2 from the atmosphere or we can say that roughly in one of the above reactors, 2t of CO2 shall be captured in a day.
- So, one reactor will capture about 730t of CO2 in a year if it runs continuously. Assuming that the reactor runs only 80% of the time, the CO2 captured will be 584t in a year.

The Projected Energy consumption in the process is given below (the numbers are on the higher side)

SL. No	Stage	Energy Consumption (KWh)	Comments
1	Calcination of Limestone	1,115	755 Mcal of energy is required to manufacture a tonne of CaO via calcination*
2	Grinding, Ball Milling & Pelleting	600#	-
3	Carbonation	208	4 fans or capacity 50K m ³ /hour of 2KW rating running for 26 Hours
4	Heating 40-70 ⁰ C during carbonation	65	2.5KW power being applied for 26 Hours
	Total Power	1,988	
	Power consumed/tonne CO2 captured using pelletized CaO with additives	2,386	Considering 20% extra power, looking at losses and other activities.

*Ispatguru – Calcination of Limestone (https://www.ispatguru.com/calcination-of-

limestone/#:~:text=Calcination%20of%20CaCO3%20is%20a,the%20carbonates%20in%20the%20limestone.) #Moraes, S. L. de, Lima, J. R. B. de, & Ribeiro, T. R. (2018). Iron Ore Pelletizing Process: An Overview. InTech. doi: 10.5772/intechopen.73164

Comparison of energy consumed per tonne of CO2 as reported.

SL. No	Company/Report	KWh/Tonne of CO2 captured
1	Climeworks	2650*
2	Carbon Engineering	2444*
3	DAC IEA	5400
4	BECCS IEA	2500
5	Wikipedia	5000
6	Novonanmek	2386

*for scaled up machines (<u>https://grist.org/technology/orca-the-largest-carbon-removal-facility-to-date-is-up-and-</u> running/)

SL. No	Energy Source	KgCO ₂ /KWh	Rs/Unit
1	Coal	0.95	11
2	Solar	0.033	2.3
3	Wind	0.009	4
4	Gas	0.45	9.5

Energy Costs and emissions related to the source of power are given below:

Source: https://www.dw.com/en/how-sustainable-is-wind-power/a-60268971

Based on the above we can calculate our energy costs based on what source of energy we use. If we use only coal-based power or gas-based power, we will not be netnegative, but as in India renewable energy may not be abundantly available, we can mix our source of energy in such a way that we are net negative. But eventually we will have to have 100% renewable energy for this process or any other direct air capture process.

Calculating the Variable Cost:

The major costs that increase proportionately with the increase in removal of CO_2 via our process are below:

	Variable Cost (Major)	INR
1	Energy (@ Rs.5/KWh*)	12000
2	Raw Materials (Limestone & Additives)	1,000
	Total (INR)	13,000
	Total (USD)	US \$156

*assuming using a proper mix of energy sources

7. Scaling up

The technology and set up as described above has the potential to be scaled up from 584t a year to 1 Million tonnes a year.

- Target: 1 Million tonnes of CO2 removed from the atmosphere
- Will require 2000 reactor units of the capacity (500t/year) as mentioned in the pilot plant section.

SL. No.	Capex & Fixed Expenditure	US \$ (M)
1	Capital Expenditure (Reactors)	10
2	Capital Expenditure (Furnaces, Ball Mills,	6
	Pelletizers)	
3	Capital Expenditure (Other)	2
4	Manpower (Operations)	8
5	Land, Office & Other Fixed Expenditure	7
6	Sales, General & Administrative	5
	Total	38

(Assumption: For this calculation, our carbon removal clock starts (for a period of 1 year) when all the machinery and manpower has been setup to remove a tonne of CO2 from the atmosphere in 1 Year.)

We need at least US\$ 38 Million in a year over our total variable cost to offset the total cost of a million tonnes of carbon capture.

In case we are removing a million tonnes of CO2 from the atmosphere, using technology as mentioned in the paper, we would need to *at least* price a tonne of carbon removal at **US\$ 194/tonne of Carbon Removed**. This would bring no profit but would only offset the cost.

With further work by increasing efficiency at every stage, some energy consumption can be reduced, but the energy consumption cannot be reduced drastically as the energy required for calcination, which is the major portion of the energy, cannot be reduced.

8. Utilization of Captured Carbon Dioxide

In this proposed process, the captured carbon dioxide is utilized by bubbling it through an aqueous solution of Lithium Hydroxide in a special reactor leading to the formation of Lithium Carbonate as per the reaction:



Fig 4: Schematic of Utilization of Captured Carbon Dioxide

The process of converting LiOH (produced from Spodumene Ore) to Li_2CO_3 by bubbling captured carbon dioxide though an aqueous solution of LiOH leads to preventing 10.8 tonnes of CO2 being released into the atmosphere per tonne of Li_2CO_3 produced.



Emission Reduction

Reference: Jarod C. Kelly, Michael Wang, Qiang Dai, Olumide Winjobi, Energy, greenhouse gas, and water life cycle analysis of lithium carbonate and lithium hydroxide monohydrate from brine and ore resources and their use in lithium-ion battery cathodes and lithium-ion batteries, Resources, Conservation and Recycling, Volume 174, 2021, 105762, ISSN 0921-3449. https://doi.org/10.1016/j.resource.2021.105762.

The reduction in Carbon Capture as shown above happens only if the LiOH is sourced from Spodumene Ore as the emissions due to production of LiOH and Li2CO2 from hard rock mines is high. LiOH produced from hard rock sources emit around 15.7t of CO2 per tonne and Li2CO3 produced from hard rock sources emit close to 20.4 tonnes of CO2. The hard rock mines are our playing ground and we can add value to the process only if LiOH sourced from hard rock mines is converted to Li2CO2 via carbonation.

Hard Rock Lithium Mines are Our Playing Ground!



Jarod C. Kelly, Michael Wang, Qiang Dai, Olumide Winjobi, Energy, greenhouse gas, and water life cycle analysis of lithium carbonate and lithium hydroxide monohydrate from brine and ore resources and their use in lithium ion battery cathodes and lithium ion batteries, Resources, Conservation and Recycling, Volume 174, 2021, 105762, ISSN 0921-3449. https://doi.org/10.1016/j.resource.2021.105762.

Fig 6: Showing emissions from LiOH & Li2CO3 produced from hard rock mines are high

The process of converting LiOH to Li2CO3 by bubbling captured carbon dioxide through LiOH has the potential to completely decarbonize Lithium mining and refinery operations. If 50% of LiOH production is converted to Li2CO3 via this process, the carbon emissions reduced is enough to offset the carbon dioxide emitted in the production of the balance 50% of LiOH lying with the refinery, thereby completely decarbonizing the mining and refinery operations of Lithium production from spodumene.



9. Economics

The equation goes this way:

LiOH (aq.) + $CO_2(g) = Li_2CO_3(g) + H_2O_3(g)$

Stoichiometrically, 1t of CO2 reacts with 0.95t of LiOH to produce 1.68t of Li₂CO₃

Average Market Price of 1t of Li2CO3 = US\$ 40,784*

Average Market Price of 1t of LiOH = US\$ 34, 250#

Our cost to remove 1t of CO2 = US\$ 156

*S&P Global Markt Intelligence, 5th July 2023 #London Metal Exchange (LME)

Gross Margin calculation considering only the major variable costs & revenue

Major Variable Costs			
Cost of 0.95t of LiOH	US\$ 32, 578		
Cost of capturing 1t of CO2	US\$ 156		
Total Variable Cost	US\$ 32, 694		
Revenue			
Revenue from Li2CO3 Sales	US\$ 65, 517		
Revenue from Carbon Removal Credits	US\$ 200		
Total Revenue	US\$ 65,717		
Gross Margin	US\$ 32, 694		
% Gross Margin (GM/Revenue *100)	50.25%		

With this kind of gross margin, the cost of Direct-Air-Capture (DAC) per tonne of CO2 captured can be completely offset while still being profitable.

10. Conclusion

We have harnessed the power of the Calcium Looping (CaL) cycle to facilitate Direct-Air-Capture (DAC), utilizing Calcium Oxide (CaO) as the solid sorbent. Through strategic innovation, we have significantly elevated the carbon capture capability of the Calcium Looping (CaL) method. This was achieved by incorporating specialized additives into the CaO powder, transforming it into a composite material. These advancements were complemented by the creation of CaO-composite powder pellets.

Our efforts yielded substantial outcomes in terms of enhanced carbon capture or carbonation of CaO. Laboratory findings demonstrated a substantial leap, with carbonation surging from approximately 65% when employing pure CaO to surpassing 95% for the composite CaO powder in its pelletized form. In our investigations, we have also discerned that a pellet shaped as a hollow cylinder would offer optimal results for DAC.

Considering economic viability, our calculations indicate that a price point of US\$ 194 per tonne of removed CO2 would effectively balance the overall expenses of carbon capture using our advanced technology. Notably, our approach boasts the potential for scalability, envisioning an expansion to 1 Million tonnes annually within a decade, necessitating a US\$ 200 Million investment.

Furthermore, our endeavours extend beyond mere capture. We propose an innovative strategy for the utilization of captured CO2, involving its passage through an aqueous LiOH solution. This process culminates in the conversion of CO2 to Li2CO3 through carbonation, successfully preventing the release of 10.8 tonnes of CO2 per tonne of Li2CO3 generated. Remarkably, our research highlights the transformative impact of adopting this approach on mining and refining operations, specifically through the decarbonization of the entire process when half of the LiOH produced from Spodumene (sourced from hard rock) undergoes carbonation conversion to Li2CO3.

An additional revelation within our work pertains to the substantial gross margin generated. This margin exhibits the potential to not only counterbalance the elevated costs associated with Direct-Air-Capture (DAC) for removing a metric tonne of carbon dioxide from the atmosphere, but to do so completely.

11. Future Work

Below are some areas where further research, development, and innovation could be focused:

1. Additive Formulation and Optimization

- Research and identify new additives that can enhance pellet reactivity, porosity, and structural integrity.
- Optimize the composition and concentration of additives to achieve the desired performance improvements.

Investigate the interaction of different additives and their synergy in promoting CO2 capture.

2. Novel Pelletization Techniques

- Explore advanced pelletization techniques, such as extrusion, spheronization, and agglomeration, to achieve even better pellet properties.
- Develop innovative methods for introducing additives during the pelletization process to ensure uniform distribution and effective incorporation.

3. Understanding Pellet Behaviour

- Conduct in-depth studies to understand how additives impact pellet microstructure, porosity, and surface area.
- Investigate pellet behaviour during the entire CaL cycle, including carbonation, calcination, and cycling effects, to optimize pellet design.

4. Process Modelling and Simulation

- Develop sophisticated computational models that simulate the behaviour of pellets in a CaL reactor under varying conditions.
- Use modelling to predict the performance of different pellet compositions, shapes, and reactor configurations.

5. Pellet Longevity and Stability:

- Investigate the long-term stability of pelletized CaO in cyclic carbonation and calcination conditions.
- Study potential degradation mechanisms, such as sintering, attrition, and the impact of cycling on pellet properties.

6. Integration with Existing Systems

- Develop strategies for integrating pelletized CaO into existing CaL-based carbon capture systems used in industries such as power, steel, and cement.
- Evaluate the economic and operational benefits of retrofitting existing systems with pelletized CaO.

7. Energy Efficiency and Environmental Impact

- Identify opportunities to further improve energy efficiency and reduce emissions associated with the process.

8. Further Study on Economic Viability and Commercialization

- Develop better business models and strategies for commercializing the technology, including licensing, partnerships, and scaling up production.

[Feedback is welcome and can be sent to arnab.sinha@novonanmek.com]