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Ice: A Review of Structure and Climate Influence

Introduction

Nearly 200 years since the first suggestion of human-caused climate change,[1] carbon emissions continue to rise.[2] Since the Industrial Revolution, humans have generated an estimated 1.5 trillion tons of carbon dioxide pollution — increasing the atmospheric carbon dioxide composition from pre-industrial levels of 280 ppm to a peak of 424 ppm this year.[3]

This excess carbon dioxide has joined with other growing greenhouse gasses such as methane, disrupting Earth's energy balance and contributing to a warming planet. This warming has summed up to an average surface temperature increase of 1.1℃ since the Industrial Revolution.[4] While there are small deviations from year to year, general trends reveal a clear increase in global temperatures. *Figure 1 shows exponential increases in both carbon emissions and global temperatures, demonstrating the strong correlation between the two.*

Though it cannot effectively account for the extent of recent emissions, the carbon cycle acts as a regulator for Earth's climate.[5] Due to the presence of carbon sinks, atmospheric carbon and consequent warming are significantly less than humans would have otherwise caused. Carbon sinks are reservoirs that absorb more carbon from the atmosphere than they release, and thus limit climate change. Sinks can be either artificial or natural, which are often divided into

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[1] *Climate Change Evidence: How Do We Know?* (n.d.). Climate Change: Vital Signs of the Planet. Retrieved November 15, 2023, from https://climate.nasa.gov/evidence
[2] *CO2 Emissions in 2022 – Analysis*. (n.d.). IEA. Retrieved November 15, 2023, from https://www.iea.org/reports/co2-emissions-in-2022
[3] *Carbon dioxide now more than 50% higher than pre-industrial levels | National Oceanic and Atmospheric Administration*. (2022, June 3). https://www.noaa.gov/news-release/carbon-dioxide-now-more-than-50-higher-than-pre-industrial-levels
[4] *Climate Change Evidence*, n.d.
[5] *The Carbon Cycle*. (2011, June 16). [Text.Article]. NASA Earth Observatory. https://earthobservatory.nasa.gov/features/CarbonCycle

two categories: oceanic, including marine biomass and carbon dissolved in seawater, and terrestrial, including vegetation and carbon within soil.[6] In the early 2000s, climate studies began to note the role of polar ice as a significant carbon sink as well.[7] Together, these carbon sinks work to balance out carbon sources such as animal respiration and human-caused emissions. *This relationship is pictured in Figure 2.*

Unfortunately, the extent of human emissions is such that climate sources prevail — leading to the aforementioned pollution and rising temperatures. The impacts of climate change are significant for both human and natural aspects of the environment.[8] Natural impacts include extreme weather patterns, biodiversity loss, ocean acidification, and land changes. Human populations feel the effects of these impacts along with the health risks of pollution and increased temperatures.

Driving Questions

Considering the severity of climate change and the relatively new discovery of ice as a carbon sink, this report is looking to explore how ice affects climate change. This will be explored from both a holistic viewpoint and through a more detailed exploration of how ice's structure allows for carbon sequestration. However, thermodynamic and kinetic principles also provide evidence for the influence of climate change on ice, so the discussion of ice as a key player in the climate must be multifaceted.

Although the Arrhenius equation $k = Ae^{-E/RT}$ explains why rising temperatures

[6] *The Carbon Cycle*, 2011
[7] Wadham, J. L., Hawkings, J. R., Tarasov, L., Gregoire, L. J., Spencer, R. G. M., Gutjahr, M., Ridgwell, A., & Kohfeld, K. E. (2019). Ice sheets matter for the global carbon cycle. *Nature Communications*, *10*(1), Article 1. https://doi.org/10.1038/s41467-019-11394-4
[8] Jackson, R. (n.d.). *The Effects of Climate Change*. Climate Change: Vital Signs of the Planet. Retrieved November 15, 2023, from https://climate.nasa.gov/effects

accelerate the rate of ice melting, we hypothesize that there is an additional factor related to changes in activation energy. Accordingly, this report will also investigate the effects of structural changes on melting requirements.

Other carbon sequestration methods will be explored as well, using knowledge of reactor flow diagrams and related mass, energy, and entropy balances.

Ice's Role

Until recently, ice was thought to be insignificant in the carbon cycle.[9] However, a 2019 study conducted by the University of Bristol led efforts to reveal ice's significant carbon sequestration potential. Compiling 20 years of research, the study highlighted the extent of two key mechanisms of glacial carbon storage: englacial and subglacial.[10]

Subglacial carbon refers to storage in the "marine sediments, soils, and vegetation" that are buried beneath ice sheets.[11] During glacial periods, this carbon surpasses that which is stored in vegetation and soil — which are typically considered to be two of the three largest carbon sinks. In fact, the Antarctic Ice Sheet alone is estimated to store up to 20 trillion tonnes of organic carbon, ten times that of the Northern Hemisphere permafrost.[12]

While the notion of significant subglacial stores originated in the early 2000s, englacial carbon storage capacity is even more recent. Atmospheric carbon becomes trapped in compacting ice as it forms, contributing to "the capacity of glacier surfaces to act as sinks for carbon-containing aerosols from anthropogenic or natural sources."[13] This also reveals the process by which the amount of carbon stored in ice is increasing due to heightened levels of

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[9] *Ice sheets impact core elements of the Earth's carbon cycle*. (n.d.). ScienceDaily. Retrieved November 15, 2023, from https://www.sciencedaily.com/releases/2019/08/190815081256.htm
[10] Wadham et al., 2019
[11] *Ice Sheets*, n.d.
[12] *Ice Sheets*, n.d.
[13] Wadham et al., 2019

carbon dioxide in the atmosphere. As more carbon dioxide is emitted, more is captured in snowfall and ice formation.

While both mechanisms allow for temporary removal of carbon from the atmosphere, it is also important to note that they create the potential for stored carbon to be released through melting. As glaciers retreat, subglacial carbon stores can become exposed and thinning ice can release methane, carbon dioxide, and particulate carbon trapped within.

Ice Structure

The discovery of englacial carbon indicates unique structural dynamics within ice. Accordingly, this section will detail ice's structure.

Ice is composed of frozen water molecules (H_2O) bonded in a hexagonal shape. When frozen, the bonds between molecules elongate, making it have a lower density than liquid water, and thus allowing it to float. When water freezes, the grains of ice will "stack" in a direction normal to the temperature gradient, which is the difference between the colder temperature, commonly the air, and the warmer temperature, typically the liquid water. This means that ice grains will typically grow vertically on the z-axis (columnar microstructure) prior to growing horizontally on the x- or y-axis.[14] *Figure 3 shows seawater normal to the freezing direction.* When each water molecule has the same orientation as the one next to it, it will be a part of a cohesive ice grain, however when the orientation shifts, deformation is caused and a new grain is formed. The grains are still connected (i.e. they are still a part of the macro ice block) and grain boundaries cannot be seen to the naked eye, however, the size and pattern of these grains have a significant impact on the mechanical, optical, and thermal properties of ice.

Larger grains are caused by slower cooling (smaller temperature gradient), whereas

[14] Cole, D. M. (2001). The microstructure of ice and its influence on mechanical properties. *Engineering Fracture Mechanics*.

smaller grains are caused by faster cooling (larger temperature gradient). Typically, ice with a microstructure with smaller, uniform grains is mechanically stronger than that with a microstructure that is inconsistent and has larger grains, as demonstrated by the Hall-Petch equation which states that $\Delta\sigma = \kappa/d$ where σ is yield strength, κ is a constant, and d is the average grain diameter.[15] *Figure 4 shows the columnar structure of lab-grown ice frozen with a 20℃ temperature gradient and small, uniform grains. Figure 5 demonstrates an inconsistent microstructure of lab-grown ice frozen with a 15℃ temperature gradient.* Smaller grains strengthen ice by obstructing dislocation movement.

Concerning to thermal properties, heat transfer in ice is completed through three separate processes: thermal conduction, radiative absorption, and convective heat transfer at the solid-gas interface. *Figure 6 illustrates heat transfer mechanisms through the ice lattice during exposure to light.* Ice also melts by radiative transfer, a mechanism that involves the absorption of light. Incident light is reflected, absorbed, transmitted, and scattered from materials. Each fractional contributor to the radiative absorption in ice sums to one ($1 = F_R + F_A + F_S + F_T$).[16] Although ice with smaller grains tends to be mechanically stronger (i.e. can withstand greater force without deformation), it also tends to melt more quickly than ice with a larger grain size as each grain has a larger surface area to volume ratio and thus is able to facilitate a faster rate of heat transfer.

Impurities

Impurities in ice are caused when non-water molecules enter the crystal lattice. Ice will rarely exist as a pure material in nature; more frequently impurities are incorporated into the ice matrix during freezing. *Figure 7 shows sea ice containing salts and extracellular polymeric*

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[15] Callister, Jr., W. D. (2009). *Materials Science and Engineering: An Introduction, Eighth Edition*. John Wiley and Sons.
[16] Callister, Jr., W. D. 2009

substances. There are two main types of impurities in polar ice: insoluble particles like mineral dust, micro-inclusions, and black carbon; and soluble ions like salts and acids that dissociate into anions and cations[17]. *Figure 8 shows microscopy of ice grains with insoluble particles. Figure 9 shows the crystal lattice of ice with a soluble impurity, methane (in blue).* Insoluble particles are not absorbed by the ice lattice as solid water is not a good solvent. Layers of ice with high concentrations of micro-inclusion insoluble impurities are called "cloudy bands", and can often be seen by the human eye.[18] *Figure 10 shows cloudy bands within ice.* These bands demonstrate well the varying distribution of insoluble and soluble impurities within ice.

Consequences

Impurities, such as CO_2 or salt, can affect the structure and characteristics of the microstructure into which they are frozen. Impurities can lie within the grains, along the grain boundaries, or across grains. *Figure 11 shows how impurity location varies in the ice matrix: within grains (red hexagon), across grains (blue circle), and at grain boundaries (green square).* Mechanical strength of materials can be affected by the location of impurities. If the impurities are across grains or at the grain boundaries they can connect adjacent domains and can inhibit crack propagation, creating a stronger microstructure.[19] Similarly, impurities at these locations act as barriers to dislocations. Grain boundaries are natural obstacles for dislocations, and adding resistance to dislocations at these locations increases strength.[20] Impurities within grains can lead to distortion of the crystal lattice. When impurities replace water molecules within the crystal structure, they disrupt the regular arrangement of water molecules, which can weaken the bonds

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[17] Stoll, N., Eichler, J., Hörhold, M., Shigeyama, W., & Weikusat, I. (2021). A Review of the Microstructural Location of Impurities in Polar Ice and Their Impacts on Deformation. *Frontiers in Earth Science*, *8*. https://doi.org/10.3389/feart.2020.615613
[18] Stoll et al., 2021
[19] Cole, David M. 2001
[20] Stoll et al., 2021

between adjacent water molecules within the grains. Insoluble impurities, such as CO_2 tend to be located in the grain interiors, meaning that carbon sequestration by ice would weaken bonds within the microstructure.[21] Such impurities interact with deformation through mechanisms such as dislocation generation, controlling grain boundary mobility, length, and energy, and recrystallization processes like grain size growth and boundary migration, all of which create a weaker crystal lattice.[22] Varying the presence of impurities in ice and the conditions under which the ice is frozen can alter the ice microstructure and thus alter macroscopic properties.

Another important aspect of CO_2 sequestration by ice is freezing point depression. Carbon dioxide lowers water's freezing point, meaning that when the concentration of CO_2 as an impurity in ice requires the temperature to be lower to freeze, consequently the ice formed will melt at a lower temperature.[23] Similarly, the density of pure ice tends to be larger than that of sea ice.[24] A higher density would encourage a stronger microstructure, as the bonds are closer together, as well as requiring more heat flow to melt, as there would be more molecules in the same amount of space. However, since sea ice has a smaller density, it will have a weaker microstructure as well as being more prone to melting than pure ice.

The combination of freezing point depression and rising temperatures leads to significant melting in polar ice. This trend is evident through the Svalbard ice core data seen in *Figure 16*, aligned peaks in black carbon concentration and annual melt percent increase indicate a strong correlation. This correlation amplifies the likelihood of significant melting, as both carbon concentration and temperatures increase — threatening further glacial thinning that has the potential to release catastrophic amounts of carbon.

[21] Stoll et al., 2021
[22] Stoll et al., 2021
[23] Arshad, M. W., Fosbøl, P. L., von Solms, N., & Thomsen, K. (2013). Freezing Point Depressions of Phase Change CO2 Solvents. *Journal of Chemical & Engineering Data*, *58*(7), 1918–1926. https://doi.org/10.1021/je3013167
[24] Ji, Q., Li, B., Pang, X., Zhao, X., & Lei, R. (2021). Arctic sea ice density observation and its impact on sea ice thickness retrieval from CryoSat–2. *Cold Regions Science and Technology*, *181*, 103177. https://doi.org/10.1016/j.coldregions.2020.103177

Solutions

Considering the limits to ice's sequestration capacity, it is imperative that other climate change solutions are found. Unfortunately, a wide-scale increase in clean technologies would not be enough to sufficiently decrease greenhouse gas emissions and consequent warming. In fact, all pathways to net-zero emissions proposed by the Intergovernmental Panel on Climate Change also require the removal of existing greenhouse gasses from the environment.[25] The two methods of this are carbon capture, which prevents the release of carbon dioxide at the source of emissions, and direct air capture, which removes existing carbon dioxide from the atmosphere.[26] The two technologies use similar methods, although preferences for chemical choice may vary slightly given different carbon dioxide concentrations. The following analysis will focus on direct air capture due to its continued potential after the green energy transition.

There are currently 27 direct air capture plants worldwide and another 130 facilities are in development, which would fulfill "the level required in 2030 under the Net Zero Emissions by 2050 (NZE) Scenario."[27] However, the process of direct air capture is also very expensive and energy-intensive, as a result of low carbon concentration and inefficiencies within the reactor flow system.

The two methods of direct air capture, which are also primary methods for other carbon capture technologies, are solid and liquid capture.[28] Solid direct air capture directs air through a solid sorbent that binds to carbon dioxide and then heats the sorbent in a vacuum to release and

[25] Levin, K. (2018, October 07). 8 things you need to know about the IPCC 1.5˚C report. Retrieved March 4, 2022, from https://www.wri.org/insights/8-things-you-need-know-about-ipcc-15c-report
[26] Meinrenken, C. (2016). Direct air capture versus post combustion capture for coal fired power plants: Energy balance and life cycle environmental assessment. *CO2 Summit II: Technologies and Opportunities*. https://dc.engconfintl.org/co2_summit2/42
[27] *Direct Air Capture—Energy System*. (n.d.). IEA. Retrieved November 13, 2023, from https://www.iea.org/energy-system/carbon-capture-utilisation-and-storage/direct-air-capture
[28] *Direct Air Capture*, n.d

capture concentrated carbon dioxide. Liquid direct air capture is a similar process through which air is passed through a chemical solvent in order to remove the carbon dioxide from the air. The solvent is a potassium hydroxide solution that reacts with the carbon dioxide to produce solid calcium carbonate. Afterward, the carbonate solids are removed from the solution and heated to release pure carbon dioxide that is then captured and stored.

To look more closely at the liquid direct air capture process, we will follow the analysis of Carbon Engineering's 1 Mt-CO2/year DAC plant as conducted by Ryan Long-Innes and Henning Struchtrup from the University of Victoria.[29] *A simplified schematic of the plant can be seen in Figure 12 and a full schematic, containing mass and energy balances as published by carbon engineering, can be seen in Figure 13.* Long-Innes and Struchtrup began their analysis with the equations $\sum M_{out} = \sum M_{in}$, $\sum NH = \sum Q - W$, and $\sum NS = \sum \frac{Q}{T} + S_{gen}$ — derived by assuming steady-state and neglecting energy changes within the mass, energy, and entropy balance equations $\frac{dN_i}{dt} = \sum(N_i)$, $\frac{d}{dt}\left[U + M(\frac{v^2}{2} + \psi)\right] = \sum N(H + \frac{v^2}{2} + \psi) + \sum Q + \sum W$, and $\frac{dS}{dt} = \sum NS + \frac{Q}{T} + S_{gen}$. Combining the energy and entropy balances, they derived the power equation $W = \sum(1 - \frac{T_0}{T})Q + \sum N\left(H - T_0 S\right) - T_0 S_{gen}$. In order to calculate net work loss, the $\sum N\left(H - T_0 S\right)$ term from the power equation was applied for all flows in and out of the entire plant. Flows in include methane, water, and air to the contactors, air separation unit, and turbine whereas flows out include carbon dioxide and depleted air from the contactors and air

[29] Long-Innes, R., & Struchtrup, H. (2022). Thermodynamic loss analysis of a liquid-sorbent direct air carbon capture plant. Cell Reports Physical Science, 3(3), 100791. https://doi.org/10.1016/j.xcrp.2022.100791

separation unit. Using the mass flow rates for *the system inflows and outflows detailed in Figure 14* and the specific enthalpy and entropy values for each given condition, the lost work sums to 258 MW. Considering a reversible work requirement of 21.2 MW and evaporative loss of 6 MW, the efficiency becomes $\eta = \frac{W_{rev}}{W} = \frac{21.2}{258+21.2-6} = 7.8\%$.

In order to investigate the causes of such inefficiency, we attempted to model the system in Aspen Plus. Unfortunately, the complex system required further developed modeling skills for a full analysis. However, building the model *as seen in Figure 15* provided a more detailed view of the components within each step.

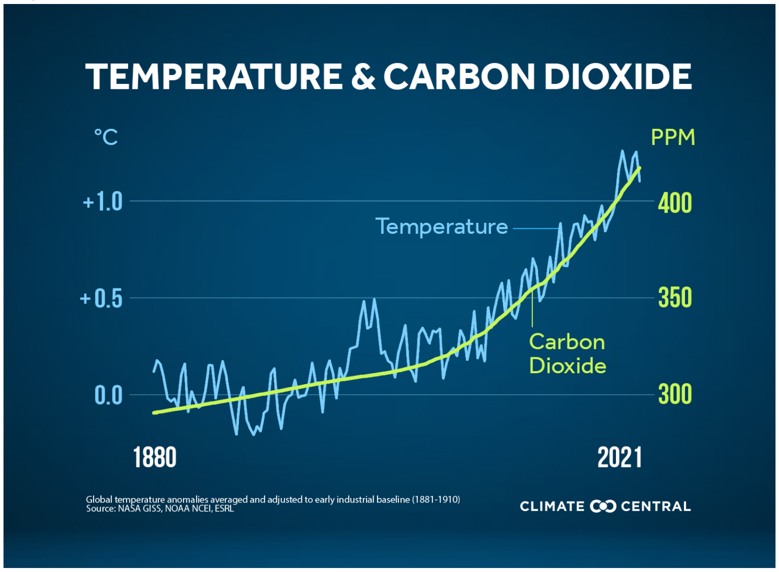
The first step, the air contactor, was modeled with a mixer, to combine the CO_2 and KOH to create K_2CO_3, and a separator, to remove the byproduct gasses. The subsequent pellet reactor was modeled with a pump and heater to prepare for precipitation, a batch reactor for $Ca(OH)_2$ addition, and a filter for precipitate removal. Next, the steam shaker was modeled with two heaters and the calciner was modeled with a stoichiometric reactor combining $CaCO_3$ and O_2, a separator, and a series of heaters for CO_2 capture. The CaO byproduct was also fed back into the pellet reactor through a recycle stream. Finally, the CO_2 was returned to the steam slaker, modeled with additional heaters leading to a separator and a multistage compressor that modeled the compression system.

Conclusion

This report has explored the role of ice as a carbon sink and its implications for climate change. Although ice acts as a significant carbon sink through englacial and subglacial sequestration of CO_2, it has a limited capacity. Rising temperatures simultaneously increase the levels of carbon dioxide trapped in ice while also accelerating the ice's melting, creating a harmful positive feedback loop, as melting glaciers and ice sheets release stored carbon. To avoid this, alternative methods of CO_2 removal are necessary. Although carbon capture systems exist and more are actively being constructed, as of right now, the efficiency of these systems can be significantly low. The DAC plant we reviewed has a calculated efficiency of only 7.8%. Improving process components to increase efficiency will be necessary for direct air capture to sufficiently scale to the level required to counteract current emissions. Creating positive headway in managing climate change requires innovation on two fronts. New technologies such as direct air capture need to continue to develop so they can actively decrease atmospheric carbon levels at a higher efficiency than they are currently able to. Similarly, it is necessary to limit further emissions and slow the melting of ice sinks, so as to not continue to exacerbate the issue. This report provides an initial glimpse into the complex relationship between ice, carbon, and climate. Further research will continue to uncover critical insights necessary to address the global crisis.

Appendix

Figure 1[30]



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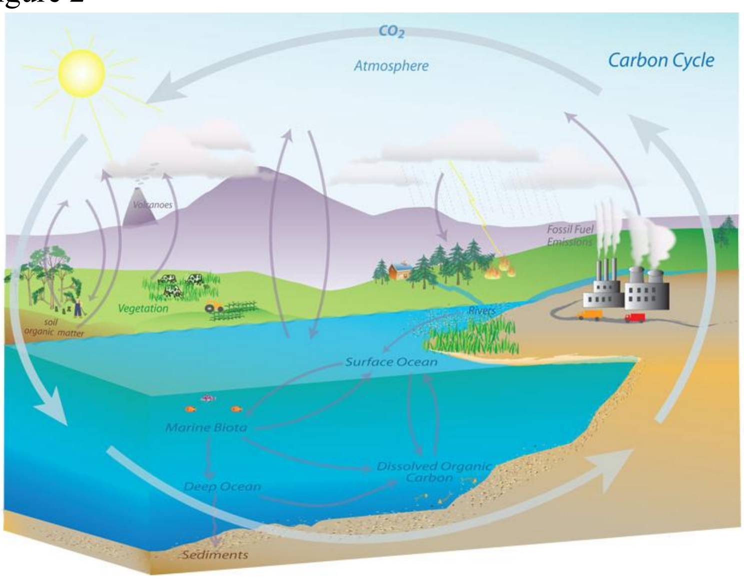

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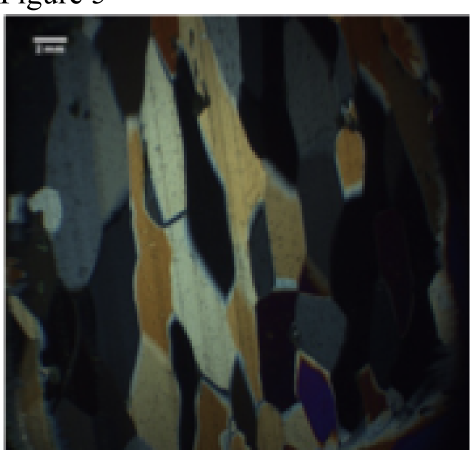
[30] *Peak CO2 & Heat-trapping Emissions | Climate Central*. (n.d.). Retrieved November 15, 2023, from https://www.climatecentral.org/graphic/peak-co2-heat-trapping-emissions?graphicSet=Annual%20CO2%20Peak%20and%20Temperature

[31] Wadham, J. L., Hawkings, J. R., Tarasov, L., Gregoire, L. J., Spencer, R. G. M., Gutjahr, M., Ridgwell, A., & Kohfeld, K. E. (2019). Ice sheets matter for the global carbon cycle. *Nature Communications*, *10*(1), Article 1. https://doi.org/10.1038/s41467-019-11394-4

Figure 3[32]

Figure 4[33]

Figure 5[34]

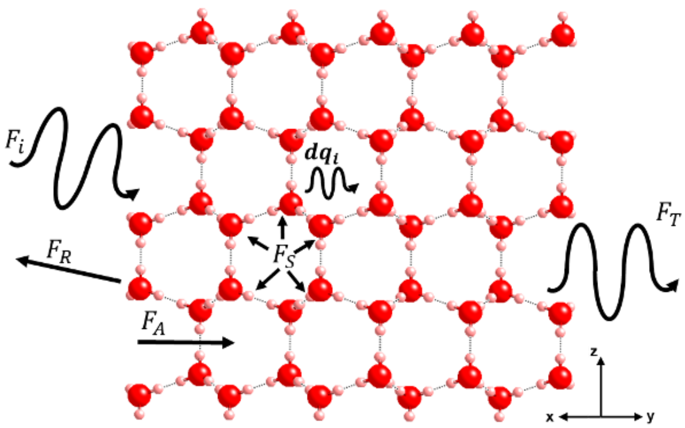


[32] Cole, David M. 2001

[33] Barry, Hayden (2022). "Reinforced Ice: Structural and Optical Characterization". USACE-ERDC Cold Regions Research and Engineering Laboratory (CRREL).

[34] Barry, Hayden, 2022.

Figure 6[35]



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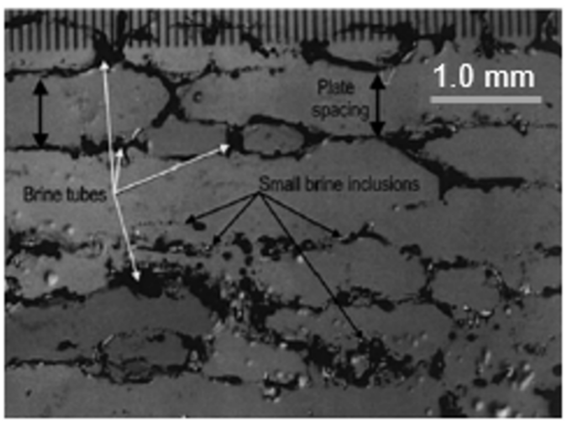

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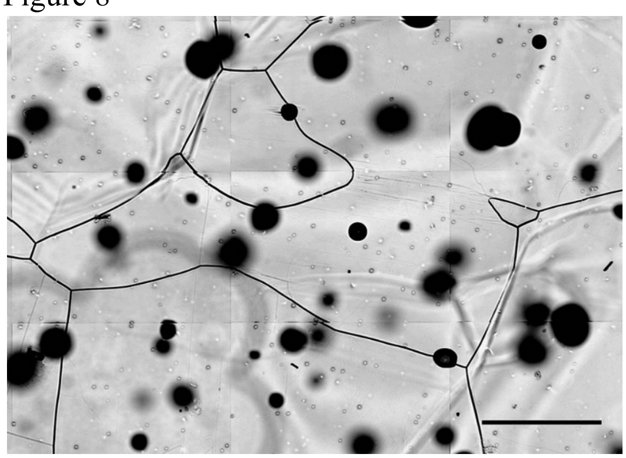

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[35] Callister, Jr., W. D. 2009

[36] Carns, R. C., Brandt, R. E., & Warren, S. G. (2015). Salt precipitation in sea ice and its effect on albedo, with application to Snowball Earth. *Journal of Geophysical Research: Oceans*, *120*(11), 7400–7412. https://doi.org/10.1002/2015JC011119

[37] Stoll et al., 2021

Figure 9[38]

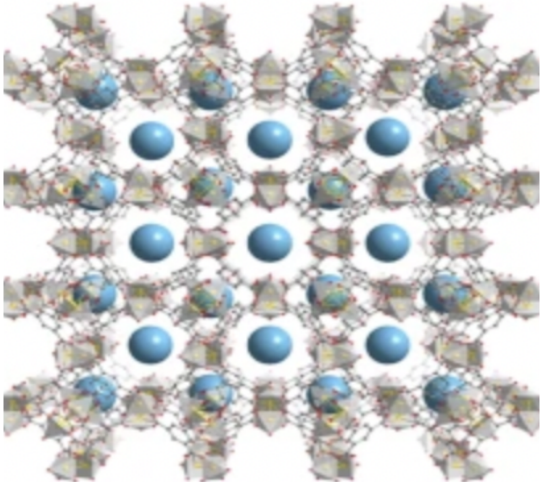
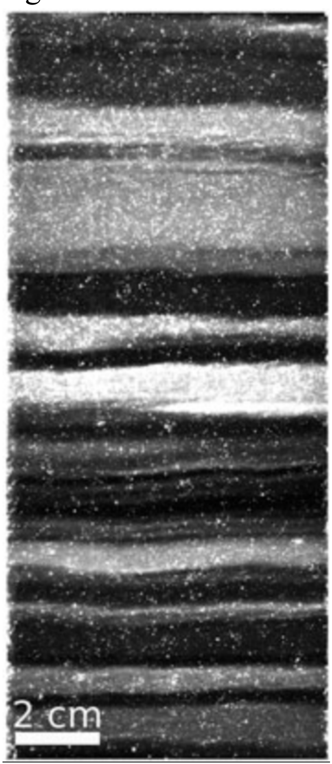


Figure 10[39]



[38] Combustible ice mimicking behavior of hydrogen-bonded organic framework at ambient condition | Nature Communications. (n.d.). Retrieved November 16, 2023, from https://www.nature.com/articles/s41467-020-16976-1
[39] Stoll et al., 2021

Figure 11[40]

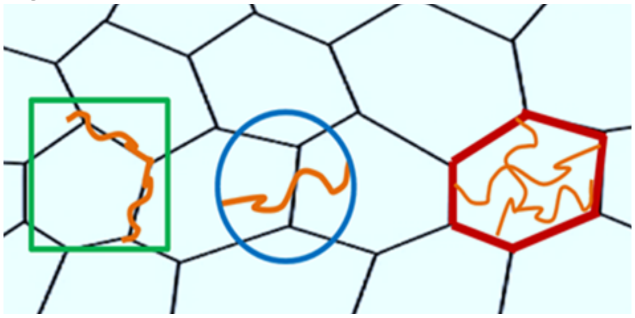


Figure 12[41]

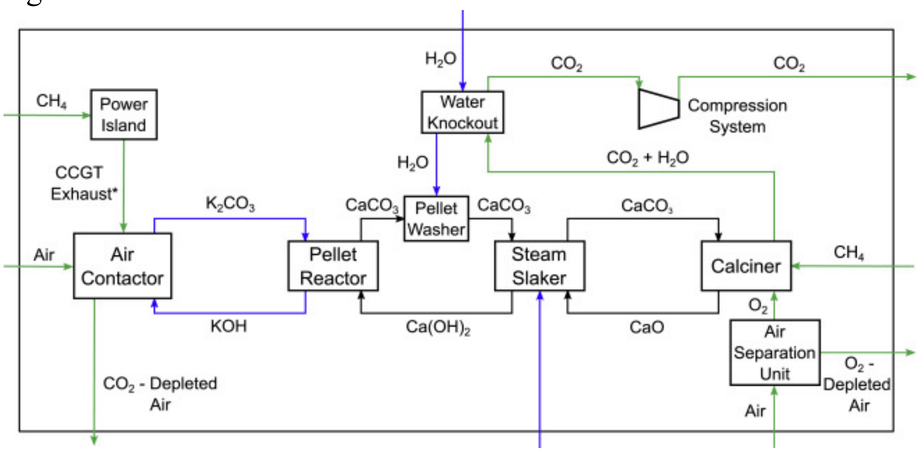
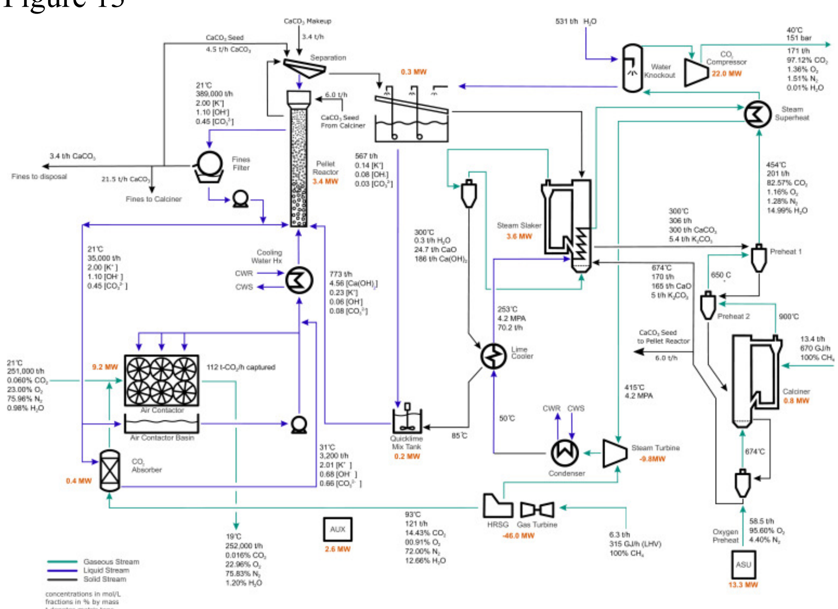


Figure 13[42]



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[40] Callister, Jr., W. D. 2009

[41] Long-Innes et al., 2022

[42] Keith, D. W., Holmes, G., St. Angelo, D., & Heidel, K. (2018). A Process for Capturing CO2 from the Atmosphere. Joule, 2(8), 1573–1594. https://doi.org/10.1016/j.joule.2018.05.006

Figure 14[43]

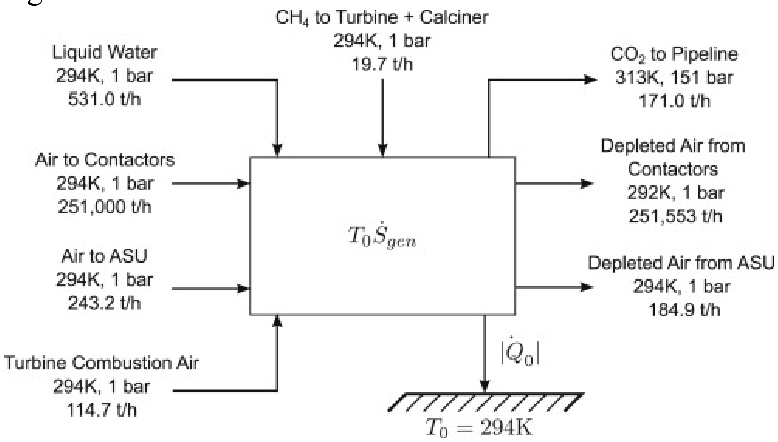


Figure 15

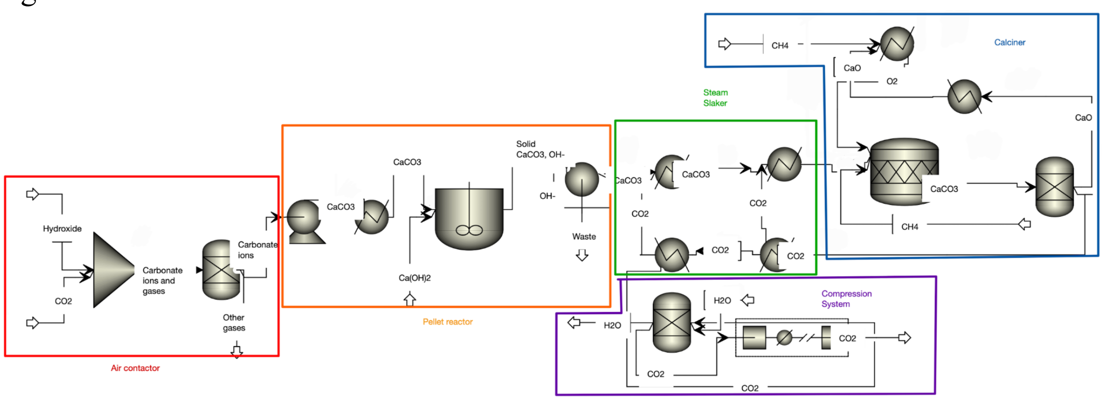
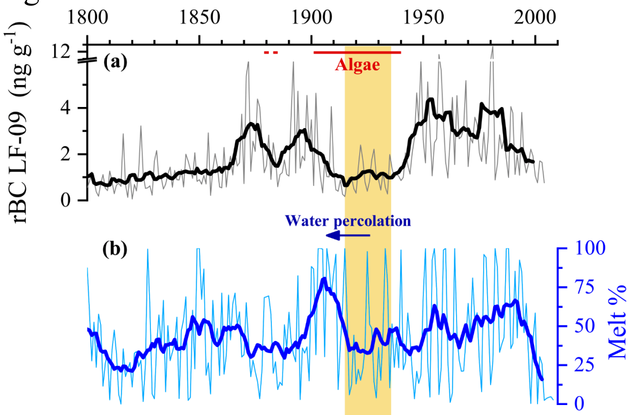


Figure 16[44]



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[43] Long-Innes et al., 2022

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