

Chapter 7

Mitigating the Climate Crisis

The global conversion from fossil fuels to renewable energy sources is a function of economic resources and political will and therefore has not been a swift process. At the same time, immediate action must be taken to eliminate carbon emissions from existing energy conversion infrastructure. This can be accomplished in a variety of ways via carbon capture and storage at the source of emissions, separating CO₂ out of the atmosphere, or replacing the fuels themselves with synthetic, carbon-neutral alternatives that existing infrastructure can use directly. In this chapter, we will look at the thermodynamics behind a few of these carbon sequestration methods and how in some cases, they can exacerbate the problem. Additionally, many major industrial processes aluminum smelting, for example, currently emit a significant amount of CO₂ as a result of chemistry underlying their operation. We will briefly look at some of these processes to understand where a significant source of emissions continue to come from and how those might be mitigated.

Finally, as a last-ditch effort in the event of emissions exceeding the IPCC targets, humanity may need to turn to major geoengineering projects to attempt to cool the Earth directly and manipulate the climate on a global scale. While considerable effort has been made towards establishing new technologies that could theoretically accomplish such a complex task, however, we are still a long way off from being able to both execute some of these strategies or even fully understand the other effects they may have on global ecosystems and weather. Nevertheless, we will apply some of our knowledge of thermodynamics to explore some of the leading concepts. At the rate global atmospheric CO₂ concentrations continue to rise, these technologies are becoming increasingly more relevant to the conversation on how to manage the climate crisis.

7.1 Artificial Carbon Capture

The material developed in Chapters 3 and 4 shows us that a) the oxidation of hydrocarbons and other carbon-based fossil fuels results in the emission of CO₂ and various other greenhouse gases as direct products of combustion reactions and b) due to the entropy generated in the mixing of these gases with atmosphere, there is mandatory work associated with their subsequent separation and capture. Because of point b) here, there are significant energy-saving benefits to capturing the gases as they are emitted and before they mix with the rest of the atmosphere. While this practice would help reduce point-of-use emissions going forward once implemented, there would still be significant emissions unaccounted for along the pipeline from fuel extraction to use. Also carbon capture at the source is often infeasible, and thus for these reasons, greenhouse gases must still be captured directly from the atmosphere as well. Both of these climate change mitigation strategies are discussed further here.

7.1.1 Carbon Capture and Storage (CCS)

We showed in Chapter 4 that the minimum amount of energy required to separate a component gas of mole fraction, X_i , from a mixture of various other gases is given by

$$w_{min} = -RT \left[\ln X_i + \frac{1 - X_i}{X_i} \ln(1 - X_i) \right] \quad (7.1)$$

per mole of component gas i , where R is the ideal gas constant, and T is the temperature at which the separation is carried out. Fig. 4.18 shows this function plotted over a component mole fraction range of 0 to 1, illustrating that the amount of energy required to separate a single gas from a mixture decreases to zero as the concentration of that gas increases to 1. The second law efficiency (i.e. w_{min}/w_{actual}) for typical separation plants is on the range of 5-40% [1], further compounding this issue. Avoiding this challenge altogether is the foundation of *Carbon Capture and Storage (CCS)* methods that remove CO₂ from the point of production where concentrations are significantly higher than in the atmosphere. It is also important to note, however, that power plants with CCS implemented incur an efficiency penalty, so the favorable thermodynamics associated with the separation become less so.

CCS methods specifically involve separating CO₂ from other gases at the points of generation (e.g. power plants and concrete manufacturers), compressing it to a liquid, and transporting it for storage in wells underground

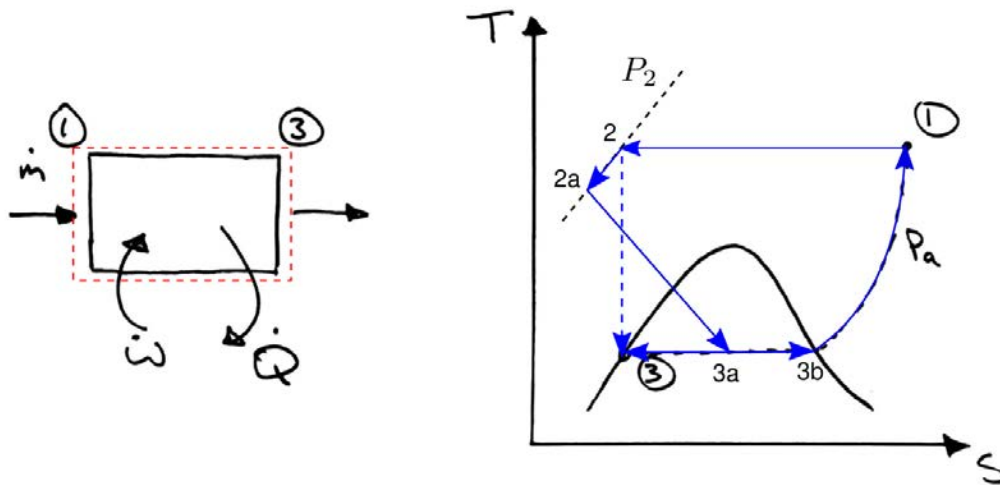


Figure 7.1: General control volume for a liquefaction process (left) and $T-s$ diagram for the ideal *Linde-Hampson Cycle* with the dashed blue line and actual *Linde-Hampson Cycle* with the solid blue lines (right).

or deep in the ocean. These solutions typically involve flowing exhaust gas with high concentrations of CO_2 past solid adsorbents like CaO and amino acid salts or liquid adsorbents like various hydrotalcites and other ionic liquids that selectively absorb or adsorb the greenhouse gas. Once separated, these sorbents can typically be regenerated using thermal energy to release the gas for subsequent compression, transportation, and storage. CCS can also be performed efficiently using polymer-based gas separation membranes to selectively remove CO_2 from the other various flue gases [2].

Gas Liquefaction

A key step that contributes to the energy requirements of CCS is the liquefaction of the captured CO_2 . This process is often necessary, as storing gaseous CO_2 is difficult and requires a prohibitively large amount of volume. As we have done before, we can compute the minimum work required for this process by formulating a general control volume and thermodynamic cycle to represent this process, as shown in Fig. 7.1. Here we see the $T-s$ diagram for two versions of the *Linde-Hampson Cycle* in which a gas at atmospheric pressure is compressed isothermally and then expanded in a number of different ways to cool and condense it into a liquid, also at atmospheric pressure.

For the ideal case, we can write the First and Second Laws for the control

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volume shown in Fig. 7.1 as

$$\dot{E}_{CV}^0 = \dot{Q} - \dot{W} + \dot{m}(h_1 - h_3) \quad (7.2)$$

and

$$\dot{S}_{CV}^0 = \frac{\dot{Q}}{T_a} + \dot{m}(s_1 - s_3) + \dot{S}_{gen} \quad (7.3)$$

Setting $\dot{S}_{gen} = 0$ for the ideal case and combining, we find that the minimum work per mass of gas converted to liquid is given by

$$w_{min} = -[(h_3 - h_1) - T_a(s_3 - s_1)] \quad (7.4)$$

which you can see is the change in availability or exergy across this cycle. The expansion process for this ideal case is represented by the dashed blue line on the T - s diagram in Fig. 7.1, indicating that the gas expands isentropically until it just intersects the vapor dome on the saturated liquid side at state 3. Here the produced liquid is also at atmospheric as shown by the fact that it lies on the same isobar as state 1 at P_a .

In reality, isentropic expansion is not possible, and thus a more realistic process is shown in the solid blue lines in Fig. 7.1. From state 2a to 3a, the compressed gas is expanded through a *throttle valve*, which holds enthalpy constant through this extremely fast expansion process¹. Because considerable entropy is generated, however, 3a typically lies inside the vapor, meaning that only some of the gas is converted to liquid. In practice, the liquid is separated and the remaining cold gas is sent back through the cycle. To save energy, this cold gas is used to cool the input gas from state 2 to 2a, bringing it back up to T_a in the process.

To determine the Second Law efficiency for this non-ideal but practical Linde-Hampson cycle, we can note that in both cases the same amount of work is required in the compression process; however, in the real case, work cannot be extracted during the expansion process as it is *isenthalpic* as previously noted. Also, not all of the gas is converted to liquid and the remaining gas must be sent through the cycle again. For this case, we can therefore compute the work required by simply dividing the work required for the isothermal compression process by the liquid quality of the outputs:

¹typically called a *flash* process

$$w = \frac{1}{1 - X_{3a}} [(h_1 - h_2) - T_a(s_1 - s_2)] \quad (7.5)$$

where X_{3a} is the vapor quality after the expansion process and thus $1 - X_{3a}$ is the liquid quality. To determine the vapor quality at state 3a, we can perform an energy balance across the *isenthalpic* expansion process from state 2a to 3a.

$$\dot{m}h_{2a} = \dot{m}[(1 - X_{3a})h_3 + X_{3a}h_{3b}] \quad (7.6)$$

$$\Rightarrow X_{3a}(h_{3b} - h_3) = h_{2a} - h_3 \quad (7.7)$$

$$\Rightarrow X_{3a} = \frac{h_{2a} - h_3}{h_{3b} - h_3} \quad (7.8)$$

$$(7.9)$$

We can simplify this further by noting that change in enthalpy of the remaining gas between states 3b and 1 must exactly be the change in enthalpy between states 2 and 2a, taking into account that there is less mass of gas after the expansion process:

$$\dot{m}X_{3a}(h_1 - h_{3b}) = \dot{m}(h_2 - h_{2a}) \quad (7.10)$$

$$\Rightarrow h_{2a} = h_2 - X_{3a}(h_1 - h_{3b}) \quad (7.11)$$

plugging this back into 7.8 and performing some algebra, we find that

$$X_{3a} = \frac{h_2 - h_3}{h_1 - h_3} \quad (7.12)$$

$$\Rightarrow 1 - X_{3a} = \frac{h_1 - h_2}{h_1 - h_3} \quad (7.13)$$

and thus

$$w = \frac{h_1 - h_3}{h_1 - h_2} [(h_1 - h_2) - T_a(s_1 - s_2)] \quad (7.14)$$

Depending on the gas, relative concentration, and its input conditions, this liquefaction process can be highly energy-intensive. For natural gas-fueled power plants, the capture and liquefaction of CO_2 can result in a First Law efficiency loss of 10-15%, requiring that more natural gas be burned to meet the demand. If the carbon dioxide can be effectively sequestered, however, this additional cost is well worth it. As we will see, however, storing the captured CO_2 is difficult and can cause additional problems.

Carbon Storage

Once compressed and liquefied, the CO_2 must be stored to prevent re-emission back into the atmosphere. The most promising methods laid out by the IPCC include mineralization, oceanic, and geological storage [3]. With mineral-based storage, CO_2 is reacted with inorganic elements to produce solid carbonates in a process similar to the natural weathering limestone to produce calcium carbonate. This enables the formation of carbonated solids that can be stored indefinitely without concern for leakage. While effective, this process can be cost prohibitive [4].

In oceanic storage, CO_2 is injected deep into the ocean where it can dissolve due to higher solubility in colder water or form heavier hydrates that sink to the bottom. There exist significant concerns, however, that the rapid increase in CO_2 and potential subsequent acceleration of ocean acidification can negatively impact marine life, ultimately causing a net-negative effect on the climate [2].

Finally, the most commonly used storage method is geological storage, in which liquid CO_2 is pumped underground into depleted oil and gas reservoirs, coal seams, or brackish aquifers. While this is the cheapest and easiest method for storing the captured gas, there can often be high leakage rates back into the atmosphere, negating its positive effects in the long term. Additionally, this method is typically used in conjunction with Enhanced Oil Recovery (EOR) and Enhanced Gas Recovery (EGR) whereby pumping liquid CO_2 into depleted oil fields enables the collection of additional oil and gas, ultimately offsetting a large fraction of the net carbon mitigation [3][2].

7.1.2 Direct Air Capture

Despite the unfavorable energy requirements given by Eq. 7.1 and shown in Fig. 4.18, it is still necessary to remove CO_2 from the atmosphere as supported by the arguments made by Solomon et al [5]. Given that molar concentrations of CO_2 in the atmosphere are around 410 ppm, the minimum work required to separate out the greenhouse gas from the air at 300 K is a substantial 500 MJ per metric ton CO_2 (140 kWh per metric ton CO_2). Regardless, many technologies are under development to accomplish this as efficiently as possible.

Direct Air Capture (DAC) generally refers to the sequestration of CO_2 from ambient air away from any CO_2 sources, where it exists in low concentrations. Many similar techniques used for CCS can also be used in DAC applications and have been seen as an ideal starting point given the significant oil and

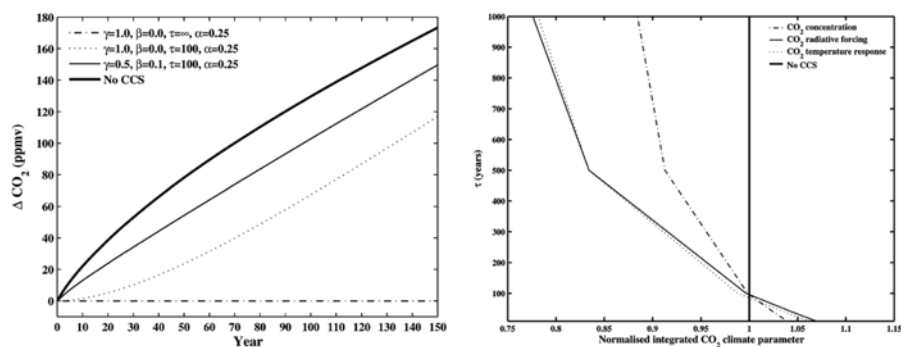
gas industry-funded development to date. Other promising approaches have been developed as well, including advanced chemisorbent materials like amine-modified mesoporous silica (e.g. TEPA-SBA-15), physisorbent materials like zeolite and metal organic frameworks (e.g. Mg-MOF-74/Mg-dobdc) [6], and ultramicroporous materials like MOOFOUR-1-Ni [7]. These materials exhibit exceptional selectivity to adsorbing CO₂ over other atmospheric gases and aerosols and can release the molecules upon heating. While promising, current limitations in manufacturing for these materials stand as a significant barrier to adoption in this space. Once the CO₂ is sequestered, this captured gas can then be stored using the previously discussed techniques. Recent research has shown that this gas can also be directly converted into alcohols like methanol to be used later as a near carbon-neutral fuel source [8].

7.1.3 Climatological Impacts of Carbon Capture

With CCS technologies, there is an inherent trade off between reducing CO₂ concentrations in power plant exhaust gases and maintaining high plant efficiencies. As previously described, work energy is required thermodynamically for any gas separation method, and for storage, additional work must be expended to liquefy and transport the captured CO₂. Even though the concentration of CO₂ is high in the power plant exhaust gases, this reduction in least work can be offset by these other factors. One of the most important factors for influencing the global climate in particular is the method of CO₂ storage and the leakage rate of the stored gas from that reservoir. Given that one of the most popular locations for carbon storage is porous geologic formations with potentially high leakage rates, this factor has a significant impact on the efficacy of CCS technologies today.

Studies have shown that for low enough gas retention times, CCS can exacerbate AGW. One study in particular found that AGW abatement is a strong function of gas retention times of the storage sites. Stone et al developed a climate model to compute the radiative forcing and resultant mean atmospheric temperature change as a function of CCS adoption rate, efficiency penalty, and residence time of the storage site, defined simply as the site storage capacity divided by the leakage rate. Fig. 7.2 shows that for a fixed efficiency penalty of 0.25, net cumulative emissions may still be significant when using storage sites with a finite leak rate, even if 100% of global carbon emissions from power plants can be captured. When looking at a 500 year analysis window, storage site residence times must exceed 100 years in order to break even with plants running without CCS [9]. The authors here additionally suggest that the use of storage with low residence times may be useful for AGW mitigation strate-

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Figure 7.2: Model results for cumulative carbon emissions over time as a function of fraction of global carbon emissions suitable for CCS, γ , fraction of emissions released by each plant into atmosphere, α , and storage residence time τ (left) and abatement of AGW causes and effects due to CCS as function of τ for 500 year analysis window (right) [9].

gies which allow near-term CO₂ concentration peaks followed by stabilization in order to reduce the total cost of CO₂ reduction [10]. These strategies have shown to be risky, however, as they could potentially result in drastic overshoot of target stabilization levels. Stone et al suggest that short term storage underground may provide a less risky alternative to this strategy, though this approach has yet to be validated.

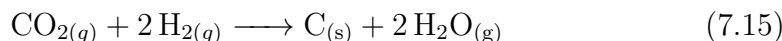
DAC approaches to AGW mitigation are virtually the same as CCS from a climatological perspective. It is important to note, however, that the costs of DAC are significantly higher than that for CCS given that concentrations of CO₂ in the atmosphere are roughly 300 times less than in the flue gases of power plants. At an estimated cost of \$1000/tCO₂, DAC will likely prove too expensive to be implemented in the near term unless significant government subsidies are leveraged [1][11]. More recent research has shown this price could be brought down to \$309/tCO₂ with more sustainable construction practices and reduced grid-level carbon intensity [12]. CCS, at an estimated \$80/tCO₂, is currently economically viable when used with Enhanced Oil/Gas Recovery; however, this reduces its effectiveness as an AGW mitigation strategy as more fossil fuels are extracted for subsequent burning.

7.1.4 Synthetic Fuels

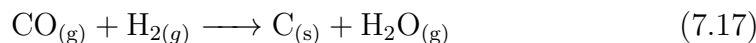
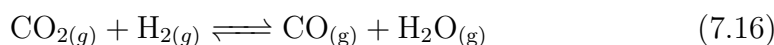
Another approach to sequestering carbon is to pull it from the air to make fuels directly, creating a net carbon-neutral fuel production system. Similar to the biofuels discussed previously, this approach involves capturing the CO₂ combustion products of various carbon and hydrocarbon oxidation reactions and recycling them into new fuel. If the additional energy input to the system is derived from a carbon-neutral renewable like solar, geothermal, or wind, the total synthetic fuel process can be considered carbon-neutral as well.

Bosch Reaction

Going back to one of the first reactions we looked at in Chapter 3 - the combustion of pure carbon in oxygen to form carbon dioxide - we discussed that the reverse reaction of decomposing CO₂ back into elemental carbon and oxygen is extremely difficult and energy intensive. A much easier reaction to carry and one that has a similar end result is the *Bosch Reaction*:



where gaseous carbon dioxide and hydrogen are reacted at high temperatures in the presence of various metal catalysts like iron and nickel to form elementary carbon and water. This total reaction is actually comprised of the following intermediate reactions:



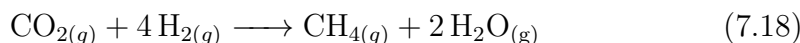
where the first is the familiar water-gas shift reaction and the second is dissociation of *syngas* into the final reaction products.

If we compute $\Delta_f h_{rxn}$ for the overall reaction at 600 °C, we find roughly 100 kJ/mol CO₂ (2.3 kJ/g CO₂) is *released* in the reaction. Indeed, this exothermic reaction actually releases thermal energy. This is largely due to the high chemical potential of the hydrogen gas reactant, and in practice, energy would be required to produce the hydrogen needed for this reaction. If this hydrogen is generated using renewable energy sources, though, this entire process is carbon negative. The carbon product can then either be stored

easily as a solid or combusted to form a carbon-neutral energy loop. As with biofuels and all carbon-neutral synthetic fuels, the primary benefit to using this process is that existing fossil-fuel combustion infrastructure can still be used with these new fuels as drop-in replacements, saving a significant amount on capital costs and making widespread adoption more likely.

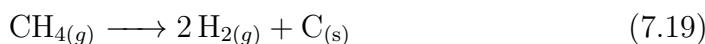
Sabatier Reaction

The natural gas equivalent to the Bosch Reaction is the *Sabatier Reaction*:



where instead of producing elemental carbon, more hydrogen is consumed - again in the presence of a metal catalyst - and methane is produced instead. For this reaction, 182 kJ/mol CO₂ (4.13 kJ/g CO₂) is released, with the thermal energy again coming from the high chemical potential of the hydrogen.

Like with the Bosch Reaction, the methane product of the Sabatier Reaction can then be combusted in existing natural gas power plant infrastructure as a carbon-neutral fuel replacement. To make this reaction a carbon-negative process, however, additional methane pyrolysis can be performed to dissociate the gas into elemental carbon and hydrogen:



This reaction requires a considerable amount of thermal energy as expected - on the order of 90.4 kJ/mol CH₄ (5.6 kJ/g CH₄k). In practice, this accomplished by bubbling methane through molten metals between 1000-1200 °C. The solid carbon product can then easily be stored or sold for processing in various other industries².

CO₂ to Methanol

Finally, carbon dioxide can be extracted from the atmosphere or hydrocarbon combustion products to produce methanol, ethanol, and other alcohols that can serve as high-density, easy-to-store liquid fuels. The reaction that produces methanol (CH₃OH) is carried out as follows:

²Numerous companies are currently using this pure carbon to generate inks and dyes used in packaging and textiles as a way to economically incentivize carbon capture.

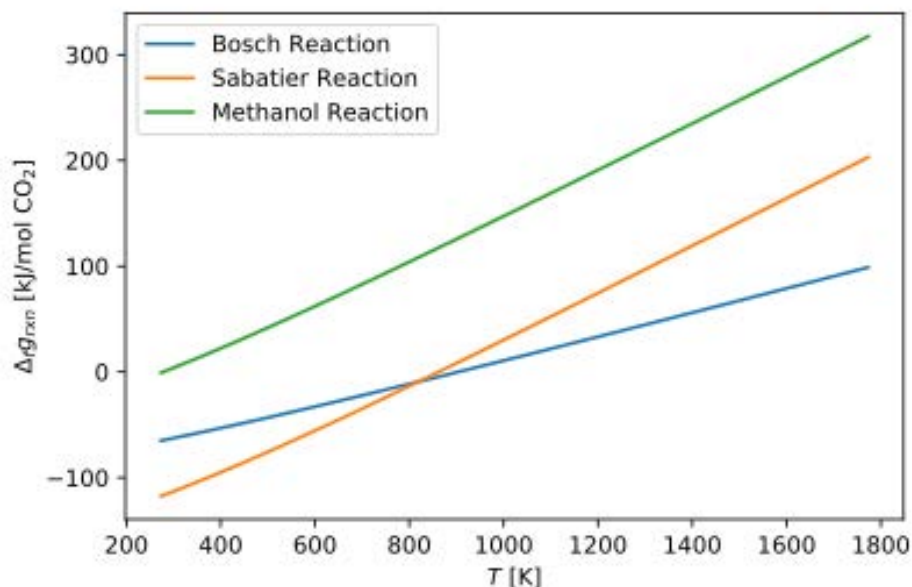
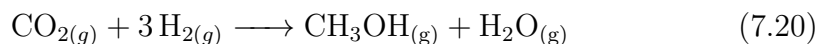


Figure 7.3: $\Delta_f g_{rxn}$ for the three carbon sequestration reactions discussed here, all at 1 bar. The lowest $\Delta_f g_{rxn}$ at a given temperature is the most favorable reaction to occur at that temperature.



where the gaseous methanol can then be condensed and easily kept as a liquid at room temperature. This reaction releases 60.4 kJ/mol CO_2 (1.37 kJ/g CO_2).

For each of these reactions presented here, the reactants include carbon dioxide and hydrogen, so a natural question that arises is: how are the product species controlled? The answer is complex and beyond the scope of this text; however, we can get a sense for which reaction is most favorable at a given temperature by finding which has the lowest $\Delta_f g_{rxn}$. As shown in Fig. 7.3, the Sabatier Reaction is favored at temperatures up to roughly 800 K, and the Bosch Reaction is favored above 800 K. The complete answer to which reaction will occur, however, requires careful control of any intermediate reactions, the concentrations of hydrogen relative to carbon dioxide, and the selection of catalysts that favor one reaction over the other [13]. Thus, even though the methanol reaction is never thermodynamically most favorable here under

standard conditions without intervention, the use of catalysts can change the picture dramatically and enable the reaction to proceed.

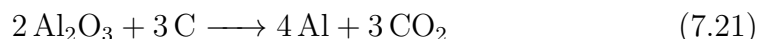
7.2 De-carbonizing Industries

As Fig. 5.15 shows, another large portion of anthropogenic carbon emissions comes from industrial processes like the manufacturing of raw materials and bulk chemicals. While there are certainly carbon emissions associated with energy required for transporting and supplying electricity and thermal energy to these processes, the bulk of these emissions are actually associated with the various chemical reactions required that produce carbon dioxide or methane as a byproduct. While there exists a plethora of different processes that fall into this category, we will only look at two here as an example of the thermochemistry involved in these carbon emissions and where we might be able to make them carbon neutral.

7.2.1 Aluminum Smelting

Globally, 64 million metric tons (Mt) of primary aluminum are produced from the reduction of Bauxite ore each year using an electrolytic process, and as shown in Figure 7.4, this value is expected to increase to over 70 Mt/yr by the end of 2020. Moreover, longer term projections show global aluminum demand increasing to roughly 150 Mt/yr by 2050 [14]. Over 25% of this aluminum produced globally goes to transportation industries including automotive, rail, and aerospace, while another 20% goes into construction materials. Packaging and electrical equipment each comprise 15% of this total production and the remaining goes into consumer products, machinery, powder metallurgy, and the deoxidation of steel [15].

Aluminum is not only one of the most widely used metals, it is also one of the costliest to produce in terms of both required energy and carbon emissions. After aluminum oxide (Al_2O_3), is extracted from Bauxite ore, it is reduced to pure aluminum via the Hall-Heroult process, in which the Al_2O_3 , is first dissolved in molten Cryolite (Na_3AlF_6), enabling the oxygen to be removed via electrolysis at carbon electrodes by the following total reaction:



which is comprised of the two electrochemical half reactions at the cathode and anode respectively:

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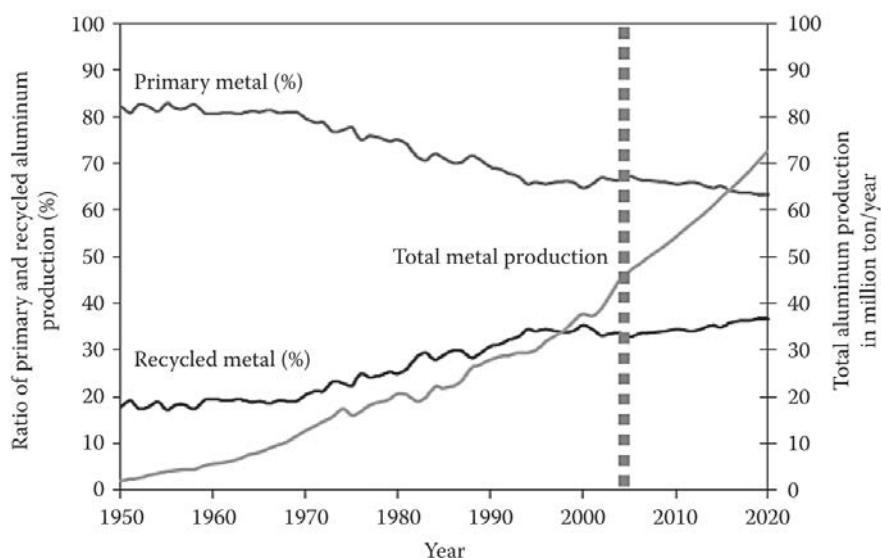
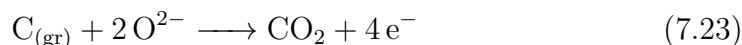


Figure 7.4: Percentage of recycled aluminum in primary production and global aluminum production rates from 1950 to the present [16].

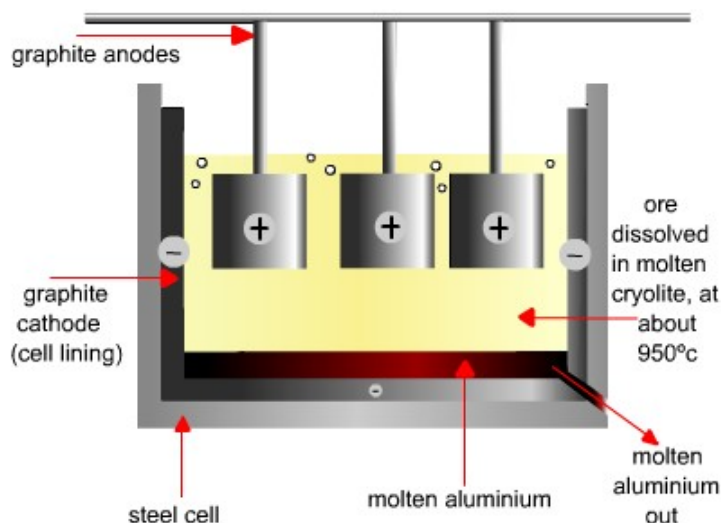
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where the aluminum and oxygen ions in each reaction are a result of the dissociation in the electrolyte. Fig. 7.5 shows a Hall-Heroult Cell that carries out these reactions.

The Hall-Heroult process requires a significant amount of energy, equal to 14,000 kWh/ton of aluminum averaged globally, and in total consumes 3.5% of global electricity production. Additionally, regardless of the carbon intensity of the electricity used to carry out the electrolysis step, as shown in Equation 7.21, this process directly results in the generation of carbon dioxide. In total, the production of aluminum results in the release of 13 tons of CO₂e per ton of aluminum, over 70% of which comes from the Hall-Heroult process alone [18][19].

Globally, this process accounts for 830 Mt of CO₂e annually, nearly 1% of the world's total carbon emissions [14]. Recycling aluminum avoids this



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Figure 7.5: Hall-Heroult Cell used in the reduction of pure aluminum from aluminum oxide. Aluminum oxide is first dissolved into a molten electrolyte and then reduced via electrolysis across graphite electrodes. CO_2 is released in the process [17].

Hall-Heroult process, significantly reducing the total carbon intensity of the resultant aluminum. Therefore, with demand for aluminum expected to increase by 130% by 2050, coupled with the increasing urgency of reducing global carbon emissions to slow the rate of anthropogenic climate change, there is significant motivation to improve the efficacy and adoption of aluminum recycling to reduce primary production.

The ultimate solution to removing carbon emissions from the aluminum production process, however, will require a move towards the use of carbon-free electrodes to perform the electrolysis. The use of an *inert anode* in this context would theoretically eliminate the carbon emissions associated with aluminum production if the electricity supplied were also carbon-neutral. The development of these anodes has been slow, as it has been challenging to find a material that promotes the aluminum reduction reaction while also not corroding in the molten electrolyte. Success has been found in using electrodes comprised of various combinations of nickel, lithium, tin, lead, copper, and cobalt [20]. Recently, Elysis, a collaboration between the major Canadian primary aluminum producers has announced the development of a production-

scale plant that will use such inert anodes, forging a path forward to de-carbonize this industry.

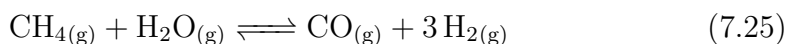
7.2.2 Hydrogen Production

The other major industry we will touch on in this section is hydrogen production. As discussed previously, the combustion of hydrogen only produces water and is thus a good candidate for storing excess energy produced by various renewable energy sources. The global demand for hydrogen far exceeds what can be produced by this excess energy, however, as it is used in many different industries, including fertilizer production, oil refining, and even electronics manufacturing. In fertilizer production, for example, ammonia (NH_3) is a critical ingredient that is produced synthetically via the *Haber-Bosch* process:

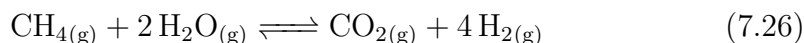


which of course requires large amounts of hydrogen.

The cheapest - and therefore most widespread - method for producing the hydrogen needed for these industries is *steam methane reforming* in which methane is reacted with water at high temperatures in the presence of a nickel catalyst to produce hydrogen and carbon monoxide:



Additional hydrogen can then be produced via the water-gas shift reaction, giving us a total reaction:



This reaction is highly endothermic, requiring a large thermal energy input of roughly 48 kJ/mol H_2 . As a result of the CO_2 released in this reaction, the production of hydrogen currently has a large carbon footprint, which is further compounded by the fugitive emissions associated with the mining and transportation of the natural gas required for these reactions. Consequently, we stand to benefit greatly from the development of new processes that reduce these emissions. Carbon-neutral hydrogen can be produced, for example, via the methane pyrolysis reaction discussed previously and given by Eq. 7.19, which produces hydrogen gas and solid carbon that can be easily filtered out

and stored. Additionally, this hydrogen can be produced directly from water via electrolysis, but currently the economics are in favor of using steam methane reforming³.

7.3 Geoengineering

Up to this point, our discussion on climate crisis mitigation has been centered on the technologies and practices that, if implemented on a global scale, would drastically limit or even eliminate anthropogenic carbon emissions. What if these solutions are not adopted or fail to work as planned? This question brings us into another realm of mitigation strategies that attempt to modify the climate and various biospheres *directly* in what is typically referred to as *geoengineering*. It has been shown that various types of direct climate intervention can have far-reaching and quick-acting effects, but as of writing this text, climate forecasting models are not powerful enough to fully model the impacts, making such interventions risky as well. Here we will discuss a few proposed strategies in this category.

7.3.1 Biosequestration

Earlier we discussed artificial carbon capture as a way to remove greenhouse gases from the atmosphere to offset emissions from industry. Another leading approach to sequestering atmospheric CO₂ involves much less technological development than the previously described methods. This approach, called biosequestration, leverages the use of natural carbon sinks to sequester the greenhouse gas via reforestation to repair ecosystems damaged by human land use and afforestation to provide additional sinks that did not previously exist. In principle, CO₂ is consumed via photosynthetic processes to form cellulose and other carbon-based plant products that store carbon in solid form. Plants, trees, and other vegetation undergo aerobic respiration and therefore emit roughly as much CO₂ as they consume in steady state operations, however, as they grow, they store more carbon in their added biomass. Additionally, as plants die, their biomass becomes incorporated into soils, giving rise to a potentially significant carbon inventory in terrestrial soils within old-growth forests. In total, these terrestrial sinks are able to sequester carbon at roughly 2.4 Pg C (9.2 Pg CO₂ equivalent) per year globally [21][22]. Pan et al estimate

³Given the abundance and ease of extracting methane, it is unlikely that the economics will work against its use unless governments enact legislations like a *carbon tax* that penalizes companies from using carbon-intensive processes.



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Figure 7.6: Mangrove forest in Gazi, Kenya showing the tortuous root system that increases carbon sequestration capabilities [23].

that when accounting for current deforestation, the net sink is 1.1 Pg C per year.

Biosequestration also encompasses the sequestering of CO_2 in marine-based ecosystems. Carbon that is captured and stored in the plant matter of mangroves, seagrass, and other marine-based plants, is referred to as “blue carbon” and serves as a significant global carbon sink. Compared to terrestrial sinks, marine-based sinks comprise a significantly smaller area; however, marine-based sinks sequester carbon at a similar rate globally. For example, carbon is buried in mangroves, salt marshes, and seagrass bed at a rate of 30 to 100 Tg C per year in each ecosystem, compared with 50-80 Tg C per year for tropical and boreal forests. In terms of global carbon sequestration rates, blue carbon sinks in mangroves, salt marshes, and seagrass beds alone have a global capacity as high as 0.2 Pg C per year. One of the primary reasons these carbon sinks are so effective is that their complex root structures, coupled with tidal inundation and tidal-driven mixing, enable them to trap and incorporate carbon-rich particles from the water into the local sediment [23]. Fig. 7.6 shows an example mangrove forest in Gazi, Kenya, which exhibits this complex root network.

Finally, biosequestration can also refer to the use of biomass to offset carbon emissions in other industries. One obvious application is to simply burn wood or methane off-gassed from the anaerobic digestion of organic matter instead of fossil fuels [24][25]. In theory, these fuels are carbon neutral, as

the carbon they release upon burning was taken in from the atmosphere relatively recently. Inefficiencies of transporting and processing these fuels results in net carbon emissions; however, these fuels may serve as a less detrimental source of energy during the transition from fossil fuels to even less carbon-intensive solar and wind energy sources. Additionally, wood may also be able to replace products that would otherwise be manufactured from aluminum or steel, whose production results in the release of significant amounts of CO₂, thereby potentially offsetting significant carbon emissions.

There are numerous beneficial feedback loops in regards to how atmospheric CO₂ concentrations affect biomass-based carbon sinks. It has long been understood, for example, that increased concentrations enable plants to grow more quickly and use less water in the process, potentially introducing a negative feedback loop to help stabilize CO₂ concentrations [26]. More recently, researches have begun to understand the more complex ways in which the Earth's forests affect the climate on the whole. Most notably, while forests provide a potentially large global carbon sink, they also have the ability to both warm and cool the climate by lowering surface albedo and increasing cooling via evapotranspiration. For example, the decrease in surface albedo due to boreal forestation in high latitudes has been shown to increase local warming, especially in snow covered regions where the forests mask the increased albedo due to snow and ice formations [27]. Globally, these forests can still provide a net cooling effect due to sequestered carbon, but with significantly reduced efficacy.

Tropical forests, on the other hand, have a greater potential to cool the planet as the decrease in albedo is more than compensated by additional evaporation rates as a result of transpiration [28]. As shown in Fig. 7.7, deforestation of tropical forests has a significant impact on global mean temperature rise, due both to their significant carbon stocks and these secondary cooling effects. In this simulation produced by Bala et al, it was also shown that the deforestation of of boreal forests results in a slight global cooling effect, further illustrating the importance of these secondary climatological effects [28]. Consequently, when considering afforestation as a means of AGW mitigation, it is important to consider not only how much land to convert or restore, but also where.

Marine-based biosequestration also has additional benefits and drawbacks from a climatological perspective. First, mangroves and seagrass beds help to prevent coastal erosion, which if left unabated can accelerate the leaching of organic carbon and ultimately reduce the carbon sequestration capabilities of those ecosystems. It has been shown that clearing mangrove forests, for example, can result in a 50% decrease of sedimentary carbon stocks over an 8

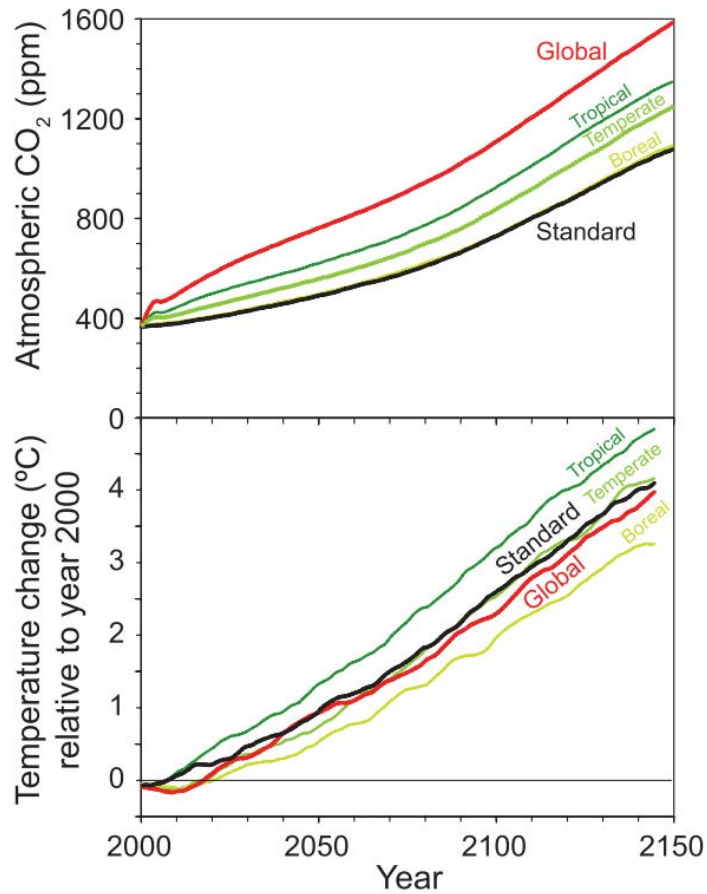
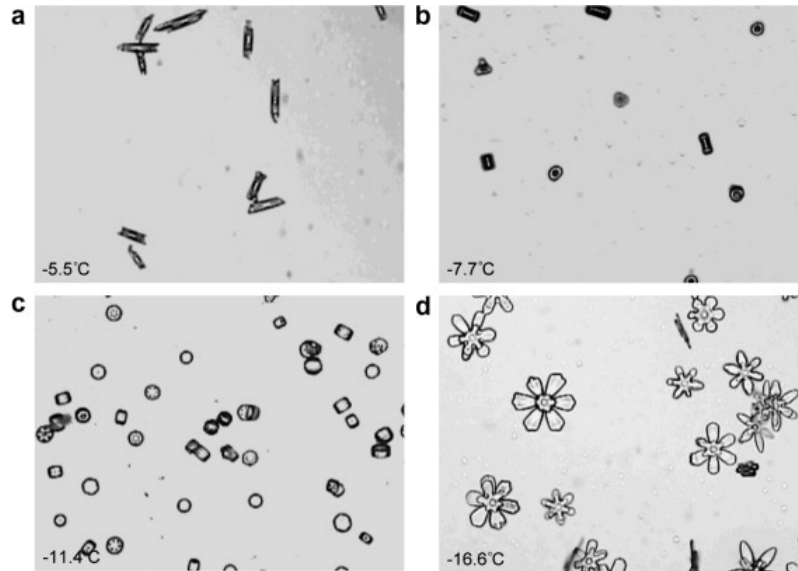


Figure 7.7: Effect of deforestation on global atmospheric CO₂ concentrations and mean temperature rise broken out by forest type [28].
Image courtesy of G. Bala, K. Caldeira, et al. "Combined Climate and Carbon-cycle Effects of Large-scale Deforestation." *PNAS*. 104 (16) 6550–6555, 4 2007



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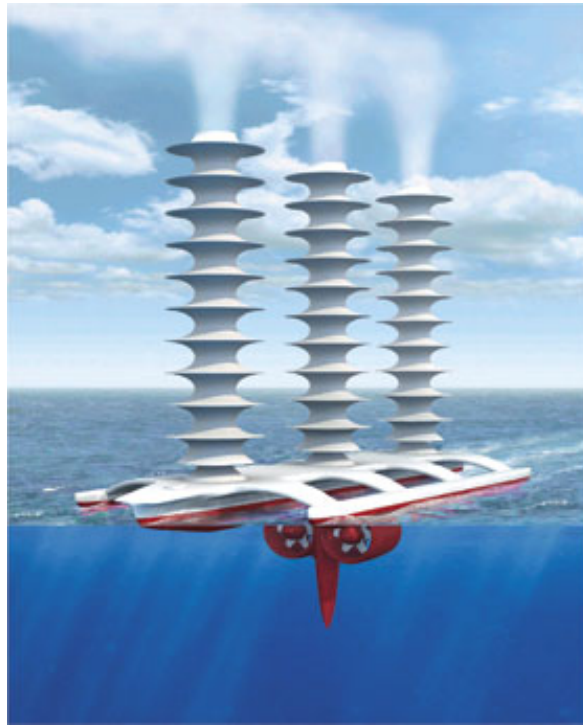
Figure 7.8: Nano-scale silver iodide particles acting as nucleation sites for water crystal (ice) growth [31].

year period [23]. Additionally, mangroves in particular are resilient to changes in sea level, making them a potentially ideal tool for mitigating AGW [29]. Counter to these positive effects, it has been shown recently that mangrove forests emit methane in rates that can reduce their global warming offset potential by 20% globally [30].

7.3.2 Cloud Seeding

Going back to Chapter 2, we learned that the driving mechanism behind climate change is the greenhouse effect, and up until now, we have primarily been focused on mitigation strategies that reduce the amount of greenhouse gases being emitted from anthropogenic sources. What if instead we could limit the amount of sunlight that arrives at the Earth's surface? Indeed, if we can increase the Earth's *albedo*, more sunlight will be reflected back into space and the energy input term in our First Law equation will be reduced. As a result, the total Earth-climate thermodynamic system will experience a drop in temperature to equilibrate to the reduced solar energy input. This theory is the driving justification for *cloud seeding*, in which bright white clouds are used to reflect sunlight.

This solution may seem counter-intuitive, as in previous chapters we dis-



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Figure 7.9: Concept for using a ship to seed clouds over the open oceans [32].

cussed the fact that water vapor is a potent greenhouse gas itself, and therefore if clouds are made of water vapor, would adding clouds to the atmosphere exacerbate the issue? The key to making cloud seeding an effective climate change mitigation approach is that the water vapor is already in the atmosphere and that by adding *seeds* or more specifically, *nucleation sites*, that water vapor will form droplets and or crystallize, forming a bright cloud. As Fig. 7.8 shows, certain aerosol-based seeds like nano-scale silver iodide particles are capable of promoting water crystal (ice) growth, which in turn can yield large clouds capable of reflecting a significant amount of sunlight. At a large scale, these silver iodide particles or similar could be injected into the atmosphere using planes, ships, or other ground-based generation systems. Fig. 7.9 shows an example of a ship that could be used to seed clouds over the open oceans. Eventually, precipitation will remove these particles from the atmosphere and they will need to be replaced.

While current climate models are unable to predict the total global effects of cloud brightening and seeding or any mitigation strategy that generally at-

tempts to increase the Earth's albedo, they do indicate that on average, these practices have the ability to reduce many of the negative effects associated with climate change. Specifically, most models point to the strong trend that reducing solar radiation results in fewer and less powerful hurricanes and typhoons [33][34], as well as reducing the rate of sea-level rise [35], buying humanity more time to reduce and eliminate carbon emissions. Critics of such practices claim that while storm intensity is reduced in some regions, other regions may also see increased storm activity or even drought as a result, which would exacerbate the negative impacts of climate change for some [36][37][38], though recent studies have shown that magnitude of these negative impacts are not as significant as was previously thought [39]. Regardless, it is highly important that the global impacts - positive and negative - are well understood to ensure that climate change is not just being mitigated for those who can afford to implement the solutions, pushing the negative impacts off onto those who cannot ⁴.

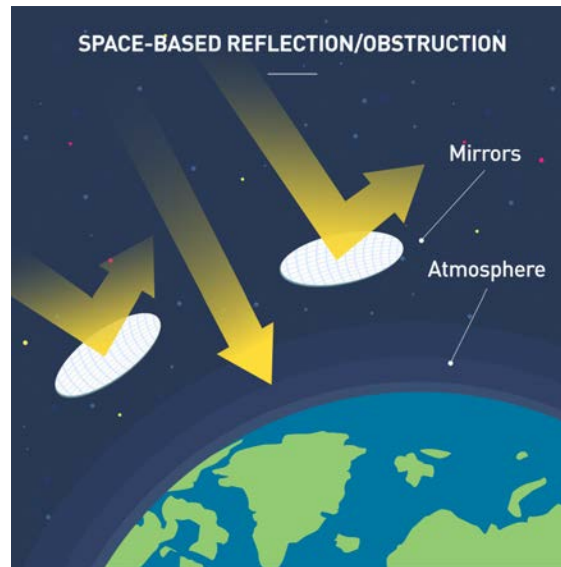
7.3.3 Other Future Strategies

The two strategies discussed thus far for using nature-based interventions to slow the climate crisis are arguably the most thoroughly researched and developed, and they also have the greatest potential to be implemented on global scale due with the necessary geopolitical and socioeconomic will. There are a few other interesting concepts to discuss briefly, though they are likely farther away from being implementable in the near future.

Space Mirrors

First, following the motivation for cloud seeding and brightening, space mirrors - mirrors attached to a fleet of controllable satellites in orbit around Earth - have also been suggested as a means of effectively increasing the albedo of the Earth thermodynamic system. As illustrated in Fig. 7.10, these mirrors simply reflect incoming sunlight to reduce the total solar flux that arrives at the Earth's surface, thereby limiting the greenhouse effect. These satellite mirrors could be placed precisely in *geosynchronous* orbit to limit the solar flux in certain regions - much like in cloud seeding - or placed to always be between the

⁴Another book could (and should) be dedicated to the ways in which thermodynamics and the resultant climate change have disproportionately negatively impacted lower-income and non-white communities and the reader is encouraged to explore the IPCC report [3] for more specifics.



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Figure 7.10: Concept for using mirrors in orbit around Earth to reflect incoming sunlight, reducing the solar flux to the Earth's surface, thereby slowing climate change [source].

sun and the Earth, casting an ever-moving shadow. In either case, these mirrors have the benefit of not needing to continuously inject potentially harmful aerosols into the atmosphere, however the logistical challenges and associated cost make this solution quite difficult to implement on a short timescale.

Making New Sea Ice

Also along the lines of increasing the Earth's albedo, another proposed climate crisis mitigation strategy is to use solar energy to produce giant ice cubes in the Arctic Ocean to combat the loss in albedo associated with melting sea ice. Fig. 7.11 shows an artistic rendering of a fleet of these ice makers producing hexagonal ice cubes at a rate comparable to the rate of ice loss. Ignoring cost and other potential barriers to adoption, we can use our knowledge of thermodynamics knowledge to perform a First Law analysis to determine if this concept is even possible.

To assess feasibility from an energy availability perspective, we can assume that the energy required to freeze these ice cubes from solar PV panels and then determine roughly how much solar panel area would be required to produce

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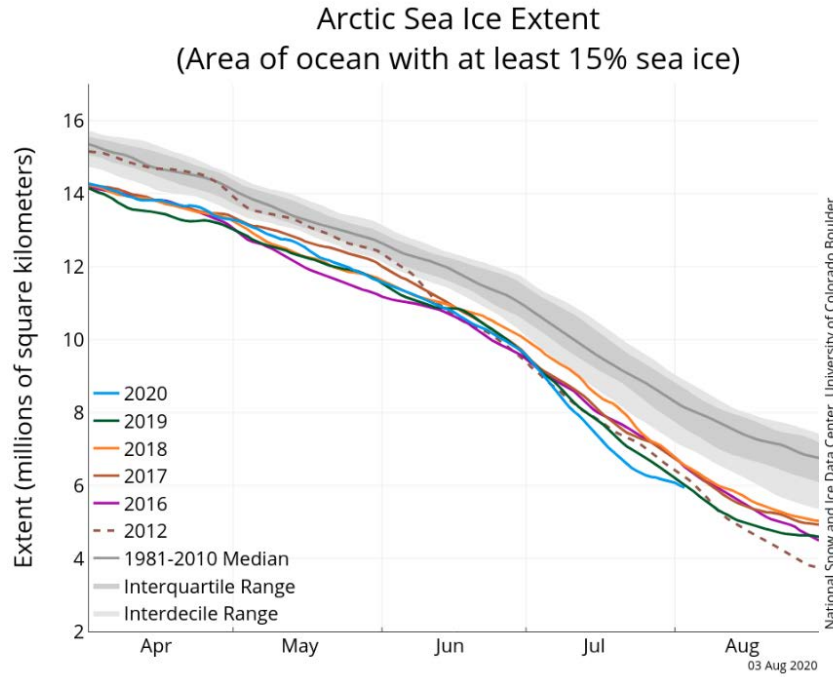
Figure 7.11: Artistic rendering of machines that could be used to replace sea ice in the Arctic Ocean to combat sea ice loss [source].

the ice that is lost each year. To begin with this analysis, we need to know first how much ice we would need to make each year. Fig. 7.12 is a plot showing the extent of Arctic sea ice and how over time, even though the values fluctuate over the course of a year, over tens of years, there is a clear average trend downwards. From this data, it is evident that the Arctic loses roughly 70,800 square km per year [NSIDC].

Assuming that the only energy we must supply is in the latent heat associated with the phase change between liquid water and ice - and ignoring the presence of the salt and other dissolved solids for now - the energy required per year is simply

$$E_{ice} = A_{loss} t_{ice} \rho_{water} h_{sf} \quad (7.27)$$

where A_{loss} is the are of sea ice lost each year, t_{ice} is the thickness of the ice cube we are using to replace the lost ice, ρ_{water} is the density of water, and h_{sf} is the latent heat of fusion of pure water. Taking the values for these variables given in Table 7.1, we can estimate the energy required to replace the sea ice per year as 2.35×10^{13} MJ, which would be roughly 4% of the total energy usage of the world. While this seems like a lot, what we are really interested in how much solar panel area we would need.



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Figure 7.12: Decline of July sea ice extent [NSIDC].

To compute the solar area, we must assume that the solar panels have some First Law efficiency, η_{PV} , the sun has an average constant solar irradiance, q''_{solar} when the sun is shining, and that the solar energy has a capacity factor - the fraction of time the sun is shining - of C . The total available electrical power we have at our disposal then is

$$\dot{E}_e = \eta_{PV} C q''_{solar} A_{solar} \quad (7.28)$$

where A_{solar} is the total solar panel area. To compute this area, we can solve for A_{solar} noting that

$$\dot{E}_e \tau_{year} = E_{ice} \quad (7.29)$$

where τ is the time period over which this analysis is performed - 1 year in this case. This gives us

Table 7.1: Variable values used in Arctic Ice Maker feasibility analysis.

Variable	Value	Units
A_{loss}	70,800	km ²
h_{sf}	334	kJ/kg
t_{ice}	1	m
ρ_{water}	997	kg/m ³
q''_{solar}	800	W/m ²
η_{PV}	0.15	-
C_{solar}	0.25	-

$$A_{solar} = \frac{E_{ice}}{\eta_{PV} C q''_{solar} \tau_{year}} \quad (7.30)$$

Plugging in numbers from Table 7.1, we find that the total solar panel area required is 25,000 km², which while considerably less than area of sea ice lost per year, is still a considerable amount of surface area. Additionally, marine environments pose engineering challenges, including corrosion and salt spray reducing the efficiency of the solar panels. Therefore, while theoretically possible, it is unlikely that this strategy will be feasible with existing technology and resources.

7.4 Summary

Capturing CO₂ via both artificial and natural methods has the potential to offset the nearly 42 Gt CO₂ emitted each year by human activity; however there are some potential factors that may limit the efficacy of these approaches. Artificial carbon sequestration, for example, relies heavily on the ability able to reliably store the captured gas. CCS technologies, by which carbon is sequestered from the output of power plant exhaust gases, may result in net additional carbon emissions over a long time horizon when used in conjunction with storage sites with high leakage rates. Renewable-driven DAC technologies avoid this effect to some degree, but their significant implementation costs (\$1,000 per ton CO₂) have proven to be prohibitive. Regardless, as fossil fuels are increasingly phased out over the next century, the total impact of CCS will become reduced as additional CO₂ emissions ideally wane. Consequently, DAC will become crucial to avoid the long-term warming trend after emissions cease as predicted by Solomon et al.

We then discussed the importance of removing carbon emissions associated with industrial processes like aluminum smelting and hydrogen production, which current rely on chemical reactions that result in the release of carbon dioxide as a byproduct. Such practices will eventually need to be converted to carbon-neutral processes to mitigate the climate crisis.

In addition to these artificial approaches, carbon can also be captured effectively and cheaply with comparatively minimal technological requirements via biosequestration. Currently, in total, terrestrial and marine-based plants and trees together take in on the order of 10 Gt CO₂ per year. Even doubling the amount of forested area would capture less than half of additional anthropogenic emissions, which on its own is insufficient from a carbon perspective; however, as previously described, the effects of afforestation on mean temperature is a strong function of the type and location of the forests. Afforestation in the tropics, for example, may have significant additional cooling benefits due to increased evapotranspiration and negligible changes in albedo.

Finally, in this chapter, we also discussed several other means of artificially increasing the Earth's albedo using cloud seeding and the more hypothetical - for now - space mirror and Arctic sea ice maker concepts. More research must be done, however, in order to determine the total impacts of these practices to ensure that the problems are not just being solved for some and exacerbated for others. Regardless, given the scale of the crisis and the difficulty in implementing many of these solutions, it is likely that the Earth will continue to warm over the next several decades. In the final chapter, we will explore how we can use thermodynamics to adapt to a climate that has already changed, helping to bridge the gap between where we are now and a sustainable world.

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