NUCLEAR POWERED CO$_2$ CAPTURE FROM THE ATMOSPHERE

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Abstract

A process for capturing CO_2 from the atmosphere was recently proposed. This process uses a closed cycle of sodium and calcium hydroxide, carbonate, and oxide transformations to capture dilute CO_2 from the atmosphere and to generate a concentrated stream of CO_2 that is amenable to sequestration or subsequent chemical transformations. In one of the process steps, a fossil-fueled lime kiln is needed, which reduces the net CO_2 capture of the process. It is proposed to replace the fossil-fueled lime kiln with a modified kiln heated by a high-temperature nuclear reactor. This will have the effect of eliminating the use of fossil fuels for the process and increasing the net CO_2 capture. Although the process is suitable to support sequestration, the use of a nuclear power source for the process provides additional capabilities, and the captured CO_2 may be combined with nuclear-produced hydrogen to produce liquid fuels via Fischer-Tropsch synthesis or other technologies. Conceivably, such plants would be carbon-neutral, and could be placed virtually anywhere without being tied to fossil fuel sources or geological sequestration sites.

Introduction

The capture of CO_2 from the atmosphere is the subject of intense research, and is viewed as a means to reduce the impact of dispersed CO_2 point sources on atmospheric CO_2 levels. Point sources such as automobile emissions and household furnaces are not amenable to direct CO_2 capture on a wide scale. Indirectly, photosynthesis and the formation of mineral carbonates perform this function in nature, but the rates of CO_2 capture by natural processes are currently exceeded by the rates that biomass and fossil fuels are being decomposed into CO_2, and the net effect is increasing CO_2 concentration in the atmosphere. Large-scale industrial processes are needed to supplement natural processes for capturing CO_2 from the atmosphere if the global carbon cycle is to be brought back into balance in regard to dispersed point sources.

Among the many indirect capture methods under study, Zeman [1] proposed one such process that relies on a closed cycle of sodium and calcium hydroxide, oxide, and carbonate transformations to capture CO_2 and concentrate it into a purified stream that is suitable for storage. This process is illustrated graphically in Figure 1. In the process, air containing dilute CO_2 is contacted with aqueous sodium hydroxide, NaOH, to form dissolved sodium carbonate, Na_2CO_3. The dissolved Na_2CO_3 so created is contacted with an emulsion of calcium hydroxide, $Ca(OH)_2$, to form calcium carbonate, $CaCO_3$, otherwise known as calcite. This reaction process is called causticization. $CaCO_3$ has much less solubility in water than Na_2CO_3 and precipitates out of solution, which allows for separation of the carbonates. The $CaCO_3$ is filtered and then dried in a steam dryer. After drying, the $CaCO_3$ is introduced into a lime kiln where it is decomposed to calcium oxide, CaO, and CO_2 is released. The CaO is converted back into $Ca(OH)_2$ using exhaust steam from the $CaCO_3$ steam dryer, and the $Ca(OH)_2$ so generated is used again to create $CaCO_3$ by reacting it with Na_2CO_3. The CO_2 released by the decomposition of $CaCO_3$ in the lime kiln is compressed and packaged for storage (or subsequent use).

The process described above is not just a theoretical construct, and key steps in the process have been demonstrated. Zeman [2] performed tests on the ability to capture atmospheric CO_2 from the atmosphere using aqueous NaOH, and then demonstrated Na_2CO_3 causticization, followed by filtration.
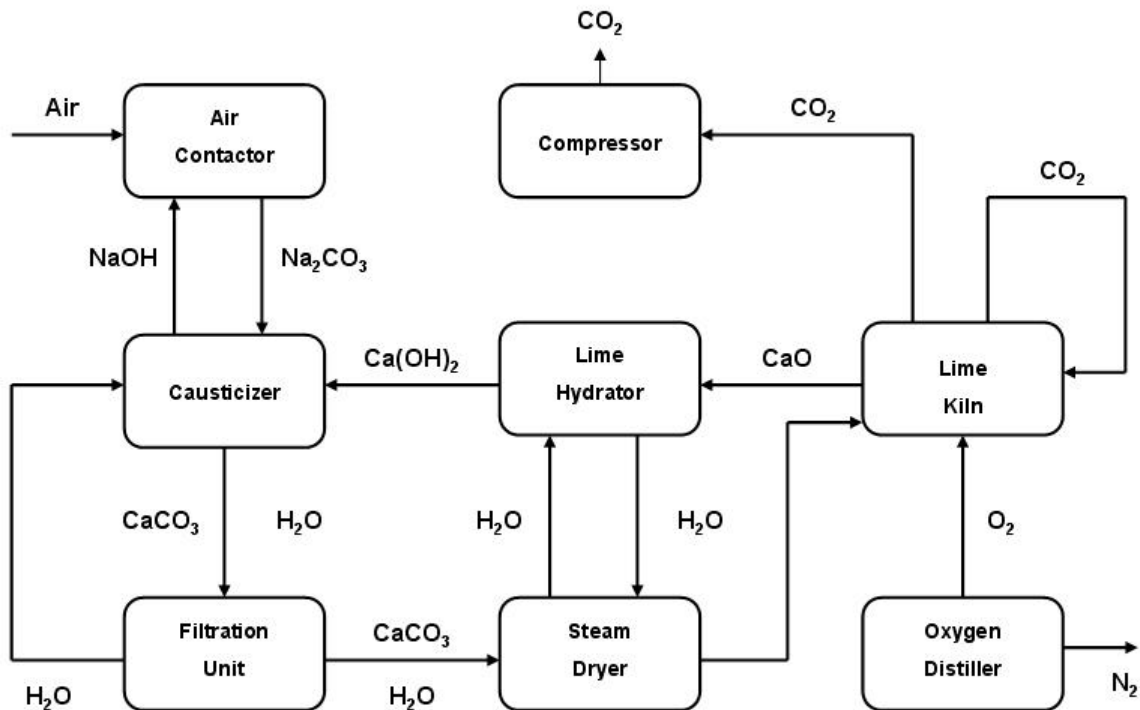
Figure 1: Atmospheric CO_2 capture process proposed by Zeman [1].

Results obtained from a series of CO_2 absorption experiments showed an average CO_2 flux into solution of 19.4 $\mu mol/m^2/s$, which translated into a CO_2 capture rate of 960 $g/m^3/h$ for the packing media used. Evaporative water losses from the air absorption step were also measured, and it was determined that about 90 g H_2O of water was lost for each gram of CO_2 absorbed. The effectiveness of the causticization reaction was measured by performing the reaction in discrete time-steps, where a quantity of solid $Ca(OH)_2$ was reacted with a Na_2CO_3 solution for a period of 10 minutes, the solids filtered and sampled, and the remaining solids contacted again with the filtrate to further the causticization process. Chemical anlyses of the samples taken from each of the steps revealed that full conversion of the solids to $CaCO_3$ occurred after three cycles with a 50% Na_2CO_3 solution, and after four cycles with a 25% Na_2CO_3 solution. Therefore, some of the key steps are achievable.

One step that was not demonstrated in the work was the use of an advanced lime kiln that uses pure oxygen instead of air to feed the fossil fuel combustion process. In commercial lime kilns, the combustion of fossil fuels such as coal or natural gas is used to heat the lime kiln to 900°C or higher,

temperatures at which $CaCO_3$ decomposes quickly into CaO and CO_2. The use of pure oxygen to enable the combustion process is suggested as an energy saving step because nitrogen would not be present in the oxygen feed stream. Nitrogen does not participate in the combustion reaction, and adds a thermal load to the lime kiln that is not productive. Also, having nitrogen in the CO_2 exhaust of the lime kiln would dilute the CO_2 produced, and additional, possibly energy-intensive, CO_2 purification steps would be needed to generate a concentrated stream of CO_2. Oxygen would be provided by an ancillary cryogenic distillation unit, which would separate oxygen from air at a relatively small energy cost (less than 4% of the total energy cost of the CO_2 capture process). Overall, eliminating nitrogen from the oxygen feed stream would improve the energy efficiency of the lime kiln, it is estimated, by up to 10%.

A drawback in the proposed process is the use of fossil fuels to heat the lime kiln. Combustion of fossil fuels such as coal or natural gas generates CO_2, and the CO_2 produced by combustion must also be captured and compressed. According to an analysis of the process [1], an additional 0.56 moles of CO_2 are produced for every mole of CO_2 captured from the atmosphere due to combustion. The net CO_2 capture of the process is still positive, because all CO_2 going through or generated by the process is captured, but generating CO_2 in a process designed for capturing CO_2 from the atmosphere is somewhat counter-productive. More CO_2 is produced as a result of the process than was originally captured from the atmosphere, and the additional CO_2 adds an energy burden to downstream compression of the CO_2 product stream. Also, if the CO_2 is headed for sequestration, a sequestration site would fill more quickly than would otherwise occur if no additional CO_2 were generated.

Using pure oxygen to enable the combustion process would improve the energy efficiency of the lime kiln, but it would not eliminate the need for downstream purification of CO_2. The combustion process does not occur efficiently if only stoichiometric amounts of oxygen are available, and excess oxygen must be provided to ensure that any fossil fuel source is converted completely into CO_2 rather than partially converted into carbon monoxide, CO [3]. Typically in air-fired fossil-fueled power plants, excess air in the amount of 10-15% is provided to achieve efficient burning of the fossil fuel. The excess air is not reacted in the combustion process and passes on through the fired section into the exhaust. For a lime kiln using a pure oxygen feed instead of air, less excess oxygen may be needed than excess

air, but some amount of unreacted oxygen may be present in the CO_2 product stream that may need to be separated or reacted in a later step.

As with any process that uses pumps and other electrical equipment, simply operating the process may also lead to increases in atmospheric CO_2 concentration, albeit marginally, if the electricity used to power the process comes from the combustion of fossil fuels in coal-, oil-, or natural gas-fired power plants, and direct CO_2 capture is not practiced at those facilities.

In order to create a more carbon-neutral CO_2 capture process, it is suggested that the additional CO_2 burden created by heating the lime kiln be eliminated by substituting the fossil fuel-fired lime kiln with a lime kiln that is heated using high-temperature heat (and possibly supplemental electricity) provided by a high-temperature gas-cooled nuclear reactor (HTGR). A nuclear-powered lime kiln would have additional benefits: the oxygen separation plant could be eliminated, and any efforts needed in purifying the CO_2 product stream for uses other than sequestration and storage would be reduced. If the nuclear plant also produces electricity, the electrical components of the process may be powered by the nuclear plant, and the CO_2 penalty resulting from the use of electricity from fossil fueled power plants could be eliminated. A description of this nuclear option is provided, and further implications of choosing a nuclear-powered CO_2 capture process from air are discussed.

Selection of Nuclear Reactor Type

Unsuitability of Using Water-Cooled Nuclear Reactors

Currently, commercial nuclear reactors throughout the world operate by heating water to generate steam, which is then used to drive turbines to generate electricity. There are variations in the type of water-cooled reactors (Pressurized Water Reactors or PWRs, Boiling Water Reactors or BWRs), but the basic process is the same. Because nuclear power plants rely on nuclear fission reactions to generate heat instead of combustion, such plants have a very low CO_2 footprint. No CO_2 is emitted as a result of nuclear fission, and CO_2 contributions only occur as a result of indirect processes (i.e., uranium mining and purification, plant construction, support operations, decommissioning, etc.). Overall, the CO_2 foot print of commercial nuclear plants is comparable, on a kWh basis, to hydroelectric, geothermal, wind,

and photovoltaic power stations, and is less than 5% of the CO_2 footprint of comparable fossil-fueled power stations [4]. Operationally, commercial nuclear pwer plants are baseload power providers and operate continuously without regard to routine weather variations or available sunlight.

A CO_2 capture plant powered by a water-cooled nuclear reactor could operate without using any fossil fuels, and no additional CO_2 would be generated during the CO_2 capture process. However, the cost of doing so would likely greatly exceed the cost of operating a fossil-fueled CO_2 capture plant because the lime kiln would require electrical heating to reach the temperatures needed ($>900°C$) to decompose $CaCO_3$ into CaO and CO_2. PWRs and BWRs operate with reactor outlet temperatures of about $325°C$ and $285°C$, respectively, and direct steam heating of the lime kiln would not be adequate. An electrically heated lime kiln would be needed, but this would require conversion of nuclear-generated thermal energy into electricity and back into thermal energy, and this can only be performed at an efficiency of approximately 33%. In comparison, a fossil-fueled lime kiln can utilize theoretically up to 100% of the thermal energy provided by combustion with thermal recuperation of the CO_2 product stream, and no intermediate electrical generation step is needed to reach the high temperatures needed to decompose $CaCO_3$. The cost of nuclear-generated electricity is currently comparable to electricity produced from coal, and there is no direct cost savings to be exploited in using nuclear-generated electricity versus direct fossil heating. If the cost penalty for producing additional CO_2 is great (e.g., carbon taxes) or fossil-fuel resources are scarce or very expensive, then it may make sense to employ a water-cooled nuclear reactor to power a CO_2 capture plant, but otherwise the fossil-fueled plant would be more economical to operate in spite of the extra CO_2 generated during operation.

Suitability of High-Temperature Gas-Cooled Nuclear Reactors

The limitations in the use of water-cooled nuclear reactors to power high-temperature processes may be overcome by using an alternative nuclear reactor type, the high-temperature gas-cooled nuclear reactor (HTGR). In an HTGR, high-pressure helium is used instead of water as the reactor coolant. With helium, there is no danger of evaporating the cooling fluid and exposing the nuclear core because the coolant is already a gas, and the reactor core is designed to use a gas coolant. Also, ceramic nuclear fuel particles embedded in a graphite matrix are used instead of metal-clad fuel, which allows the reactor

to operate at much higher temperatures without fuel failure than the fuel cladding materials used in water-cooled reactors. This replacement, along with changes in the nuclear core configuration and the nuclear vessel design, allow this reactor type to achieve outlet nuclear reactor temperatures in excess of 900°C during normal operation.

It must be noted that the HTGR concept is a higher-temperature evolution from the the Magnox and the Advanced Gas Reactor (AGR) designs still in use in the United Kingdom that use gaseous carbon dioxide as a coolant. The Magnox reactor design, which was the first generation of gas-cooled reactors to be commercialized, uses metal-clad fuel and provides an outlet temperature of no higher than 400°C [5]. The AGR is the next generation of Magnox reactor, and also uses metal-clad fuel and carbon dioxide as a coolant, but is capable of providing a reactor outlet temperature of up to 650°C. Today, only two Magnox plants and two AGR plants operate commercially in the United Kingdom.

No commercial helium-cooled HTGR facility is yet in operation, but several plants have been constructed and operated in the past. In the United States, Peach Bottom Atomic Power Station 1 [6], UHTREX [7] and Fort Saint Vrain [8] operated from 1967-1974, 1966-1970, and 1977-1989, respectively. In the United Kingdom, the DRAGON facility operated from 1964-1975 [9]. In Germany, two reactors were demonstrated – the AVR and the THTR-300 – from 1967-1988 and 1983-1989, respectively [10].

Two HTGR demonstration facilities are currently in operation: the HTTR in Japan, and the HTR-10 in China. These faciles began operation in 1999 and 2003, are sized at 30 MWt and 10 MWt, and are capable of providing nuclear reactor outlet temperatures as high as 950°C [11].

Future HTGR plants are also planned. Pebble Bed Modular Reactor (Pty) Ltd., based in South Africa, is seeking to commercialize the Pebble Bed Modular Reactor (PBMR), a design based on the German AVR, and will begin construction of the first plant in Koeberg, South Africa, in 2009. The PBMR design will be capable of providing an outlet temperature of up to 950°C, though it is unknown at this time when the PBMR design will be pushed to operate at this limit. In the United States, the U.S. Department of Energy is planning start-up of the Next Generation Nuclear Plant (NGNP) [13] by 2021, which will be capable of providing high-temperature heat at temperatures up to 950°C for generating

electricity or for providing heat to chemical process applications. A summary of past, current and future HTGR plants is shown in Table 1.

Table 1: **Past, Present and Future HTGR Plants**

Nuclear Reactor	Country	Years of Operation	Thermal Output MWt	Outlet Temp $^\circ$C
Peach Bottom Atomic Power Station 1	USA	1967-1974	115	714
UHTREX	USA	1966-1970	3	1320
Fort Saint Vrain	USA	1977-1989	842	766
DRAGON	UK	1964-1975	20	750
AVR	W. Germany	1967-1988	40	950
THTR-300	W. Germany	1983-1989	750	750
HTTR	Japan	Since 1999	30	950
HTR-10	China	Since 2003	10	950
PBMR	South Africa	2013+	400	950
NGNP	USA	2021+	600	950

A distinguishing characteristic of the NGNP from other HTGRs (aside from the HTTR in Japan), and most relevant to the proposed CO_2 capture process, is the presence of a high-temperature heat transfer interface in the proposed NGNP plant design. A high temperature heat transfer interface will enable the transmission of high temperature heat beyond the nuclear plant so that it can be used to power downstream non-nuclear applications. The transmission interface will consist of high-temperature heat exchangers, pressurized fluid conduits containing helium or molten salt, valves, and fluid motivators (i.e. pumps or compressors), and may span hundreds of meters in length. A simple schematic of the NGNP is shown in Figure 2, and an artist's conception of the NGNP connected to downstream chemical plants is shown in Figure 3. Japan's HTTR has been designed with a high-temperature thermal transmission interface also, and the thermal interface will be used (eventually) to power a high-temperature hydrogen production process that will split water into hydrogen and oxygen [14].

CO_2 Capture Process Modification

The CO_2 capture process offered by Zeman [1] may be modified to work with an HTGR by replacing the fossil-fueled lime kiln used in the process with a lime kiln that is heated indirectly by the nuclear reactor. A heat exchanger at the terminal end of the HTGR long distance heat transfer loop
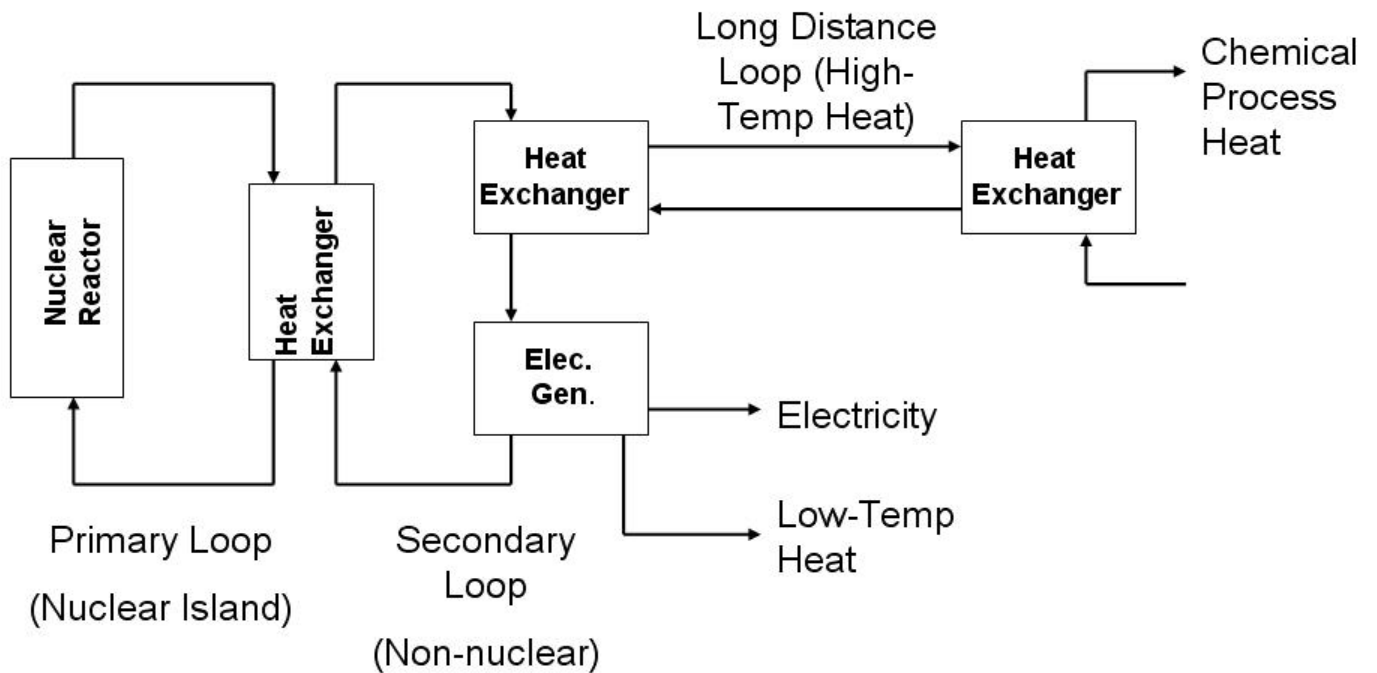
Figure 2: Schematic of NGNP with heat transfer interface.

may be used to heat a circulating stream of CO_2, and the heated CO_2 is used to heat the lime kiln. With no fossil fuel to burn, the oxygen separation plant can be eliminated. This modified process is shown schematically in Figure 4. Ideally, air is the only feed stream into the process, and CO_2 is the only product stream; all other materials are conserved in the cyclic process. In practice, make-up water will be needed to compensate for water evaporated from the air scrubbers, and some amount of NaOH or $CaCO_3$ may need to be added to make up for losses of ionic Na or Ca materials. The modified process can be further integrated with the nuclear plant if the nuclear plant provides electricity to power the electrical devices used in the CO_2 capture plant, and low-level heat from the electrical generators may be used to make steam for the steam dryer.

Some preparatory development work would be needed to adapt existing lime kiln designs for use with indirect CO_2 heating, but extensive research and development would not be needed because prototype and commercial designs already exist that could be used for this purpose. U.S. Patent 4,745,869 [15] describes a rotary lime kiln that is heated indirectly by the combustion of coal. In this invention, coal
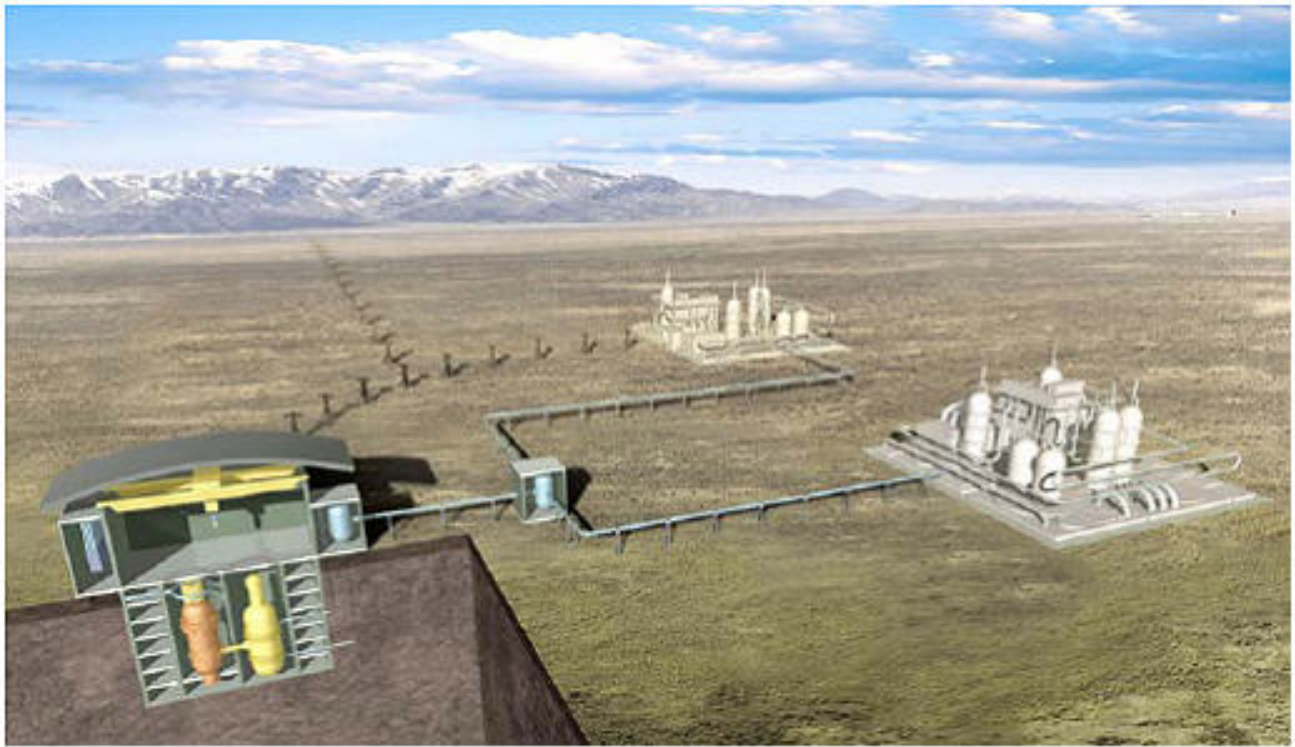
Figure 3: Artist's conception of NGNP connected to downstream chemical plants.

is combusted in a preliminary two-section chamber which is used to collect the slag and ash from burning coal, and the slag- and ash-free combustion gases, consisting principally of hot CO_2, are introduced into the rotating kiln to heat the $CaCO_3$ decomposition process. Large-scale indirectly heated rotary kilns such as the one described can be purchased on the commercial market. More recent innovations in lime kiln design have been made that would be compatible with nuclear heating, and one such invention is described in U.S. Patent 5,846,072 [16]. In this second patent, $CaCO_3$ or limestone particles are moved through a stationary ceramic tube using a screw conveyer, while the outside of the ceramic tube is heated with combustion gases. The limestone moving through the inside of the ceramic tubes is heated by contact with the tube walls, and the tube walls are heated by hot gases (or solids) that pass over the tubes. If such a lime kiln were used in the modified process, the ceramic tubes would be heated by hot CO_2 instead of combustion gases.

A disadvantage of using a nuclear heat source instead of a combustion heat source is that temperatures in excess of $900°C$ cannot be easily achieved without supplemental electrical heating. The lime kiln will operate at the temperature at which hot gas is delivered to it, and that temperature will be
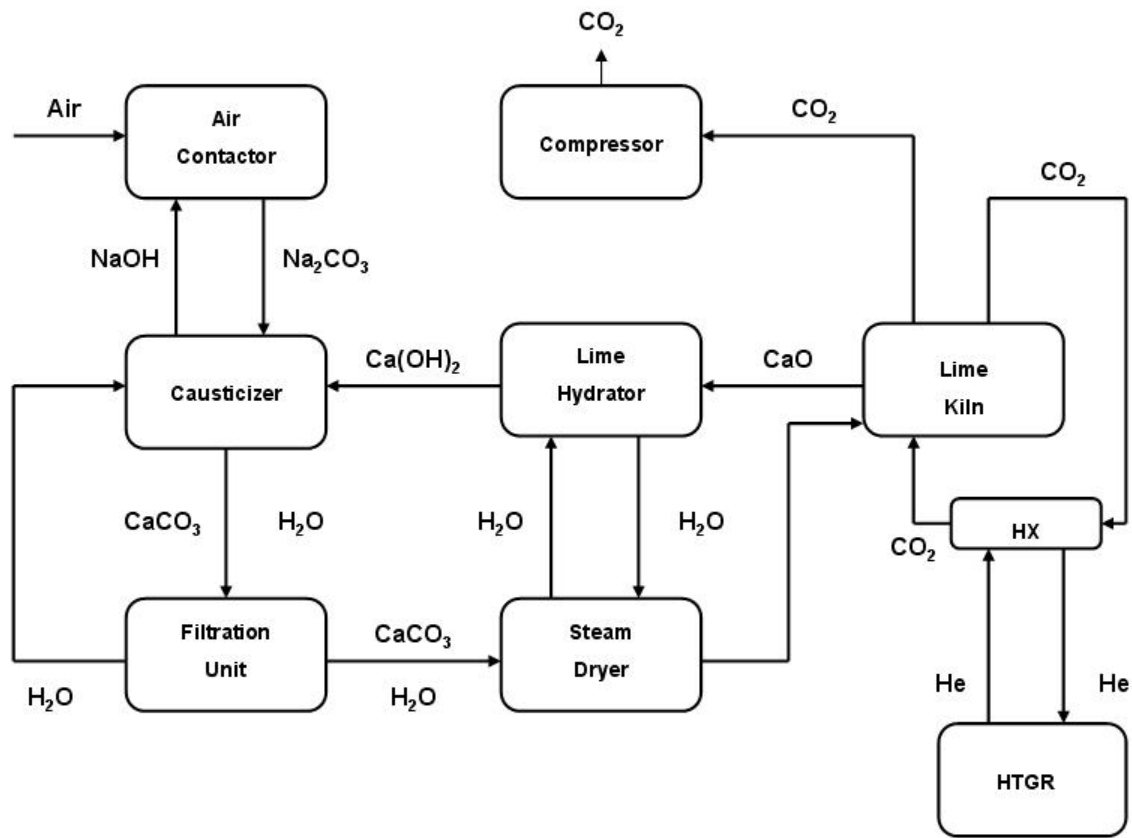
Figure 4: Schematic of nuclear-powered CO_2 capture process.

less than the outlet temperature of the nuclear reactor due to the temperature drop across intervening heat exchangers and external heat losses from the long distance heat transfer loop. This may not be a serious issue, however, because the $CaCO_3$ decomposition reaction occurs at temperatures less than 900°C, and it is more a matter of optimizing the residence time and particle size of the $CaCO_3$ in the lime kiln in order to achieve the maximum conversion to CaO and CO_2 [17] at the temperature provided by the nuclear reactor.

The kinetics of the decomposition reaction may be increased by employing supplemental electrical heating to reach a higher temperature, grinding the $CaCO_3$ to decrease particle diameter and increase surface area, or by minimizing the temperature drop between the nuclear plant and the lime kiln. Electrical heating could be done by employing a booster heater to further heat the CO_2 after it emerges from the terminal heat exchanger of the long-distance heat transfer loop, and is easily done, but this would carry a marginal energy cost penalty. Grinding the $CaCO_3$ particles to make them smaller is an option,

but this may require drying the $CaCO_3$ to full dryness, and then grinding and sieving the particles to achieve the desired particle size if the particle size of the precipitated $CaCO_3$ is larger than 100 μm in diameter [17], and that would be an additional process step. The CO_2 heat exchanger may be eliminated at the lime kiln by using an advanced lime kiln design like that described in U.S. Patent 5,846,072, and hot helium or molten salt from the long distance heat transfer loop could be used instead of recirculating CO_2 to heat the lime kiln. These are all options, and the best path for achieving an efficient process will require comparisons of process flow sheets, and capital and operating costs to determine the best option.

In areas where water is scarce, air coolers and refrigeration units may be required to recover water evaporated from the air scrubbers, and this will require additional energy input.

CO_2 Capture Process Extensions

Since nearly their inception, HTGRs have been envisioned as machines that could be used to drive, by virtue of their high outlet temperature, chemical processes that would be capable of splitting water to generate hydrogen (and oxygen). This can be done using conventional room temperature electrolysis, but water becomes easier to split into its consituents at higher temperatures, and more thermodynamically efficient and hopefully more economical processes can be developed that can effectively use the high-temperature heat provided by HTGRs. Three such processes that are currently under development by the U.S. Department of Energy Office of Nuclear Energy are the Sulfur-Iodine Process [18], the Hybrid Sulfur Process [19], and High-Temperature Electrolysis [20]. Each of these processes uses thermal energy at about 800°C or higher to split water into hydrogen and oxygen.

With co-generation of H_2, a combined H_2 production/CO_2 capture plant would provide the raw materials suitable to convert CO_2 into liquid fuels and other chemicals. CO_2 produced by the CO_2 capture plant could be combined with H_2 from the hydrogen plant to produce methanol, CH_3OH, by hydrogenation. Carbon monoxide, CO, could be formed from CO_2 by employing the reverse water-gas shift reaction between CO_2 and H_2. With CO and H_2, any number of hydrocarbons can be produced by Fischer-Tropsch synthesis, including aromatic and long-chain hydrocarbons. The chemical feedstocks

produced by such a plant could be considered "carbon neutral," in that there would be no net production of CO_2 in their production or use.

In the larger sense, combining a CO_2 capture plant with a nuclear hydrogen plant would make for a more economical plant because co-generation of useful feedstocks provides a higher value revenue stream for the plant than is likely to be achieved by collecting compensation for atmospheric CO_2 removal (e.g., carbon credits). As an added benefit, a combined plant is not geographically restricted to being near CO_2 sequestration sites, and could be located closer to markets. One can envision an integrated nuclear-powered facility that is capable of producing chemicals and fuels using only water and air as material inputs to the plant. Of course, a source of water would be needed to make hydrogen, and this could be a limitation in some arid areas. On the coastline of continents or at sea, however, a nuclear-powered hydrogen production/CO_2 capture plant could be combined with a water desalinization facility to produce fresh water to feed the hydrogen plant, and this opens up the possibility of floating fuel production plants that use only the raw materials around them to generate fuel and chemical feedstocks without burning or consuming fossil fuels.

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