CO2 EXTRACTION FROM AMBIENT AIR USING ALKALI-METAL HYDROXIDE SOLUTIONS: NICHE MARKETS TO INDUSTRIAL SCALE IMPLEMENTATION

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Introduction

Global climate change is one of the most serious environmental problems facing the world. In order to mitigate climate change, deep reductions in carbon dioxide (CO_2) emissions will be required in the coming decades. This necessity is at odds with fossil fuels' overwhelming dominance as an energy source. By capturing and storing CO_2 in sinks other than the atmosphere, atmospheric CO_2 levels can be stabilized or lowered while fossil fuels and their associated infrastructure are phased out more slowly and thus at lower social cost. Given the extent of emissions reductions needed to stabilize atmospheric CO_2 concentrations and the inertia involved in shifting the world's primary energy sources, carbon capture and storage (CCS) will likely constitute a substantial share of emissions reductions in the near- to medium-term.

Nearly all current research on CCS focuses on capturing CO_2 from large, stationary sources, such as power plants, as it is produced. Such plans usually entail capturing CO_2 from flue gas, compressing it, and transporting it via pipeline to a sequestration site. In contrast, the research proposed here focuses on capturing CO_2 directly from ambient air. The capture unit can be located at a favorable sequestration site, avoiding the need for a CO_2-transportation infrastructure. This strategy has the advantage that CO_2 emissions from any sector can be captured: power plants which are difficult to retrofit, point sources in locations where new infrastructure is expensive or impractical to build, and diffuse sources such as automobiles.

Previous research by our group indicated that dilute aqueous alkali-metal solutions derived from CaO- and MgO-rich waste streams (e.g. steel slag and concrete waste) effectively removed CO_2 from ambient air. The economic of CO_2 extraction from air using these wastes can be favorable under certain conditions, but the mass of carbon that can be captured and sequestered is small. The aim of this research is to design and analyze an industrial scale system for capturing CO_2 from ambient air that uses well-understood technologies available today, that is simple and inexpensive, and that is scalable to handle a significant fraction of anthropogenic CO_2 emissions. With these goals, the scheme depicted in Figure 1 has been devised as a basis for analysis. In it, air is forced through a cooling-tower-like structure where an aqueous sodium hydroxide (NaOH) solution is sprayed and collected by gravity. The NaOH droplets absorb CO_2 from the air, yielding $NaHCO_3$(aq). The collected solution is piped to a batch reactor for a sodium-calcium exchange process. The regenerated NaOH is recycled to the contactor and the $CaCO_3$ formed is sent to the Calciner. Upon heating the $CaCO_3$, CO_2 is driven off in a pure stream and pressurized for sequestration. The CaO formed is then recycled to the Na-Ca exchanger.

The Na-Ca exchange process is well-understood and currently used at industrial scales in the pulp and paper industry with almost the precise parameters required for this scheme (Adams, 1989). The calcining (regenerating the CaO) and hydroxylation steps are also well-understood processes used at industrial scales in the pulp and paper industry (*ibid.*) and during cement manufacture. The contactor is the least-specified component of the system and, as such, the subject of this research. A diagram of the proposed contactor is shown in Figure 2. The design is feasible in principle – CO_2 capture at ambient concentration with NaOH solution has been demonstrated

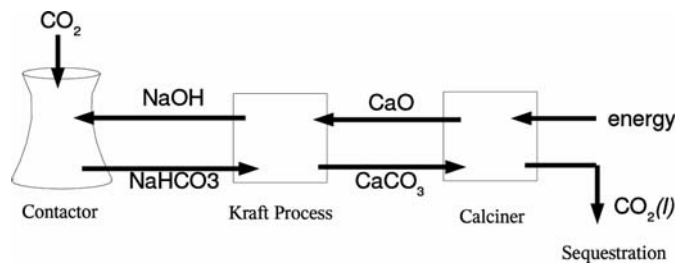

Figure 1: Proposed process for capturing CO_2 from ambient air.

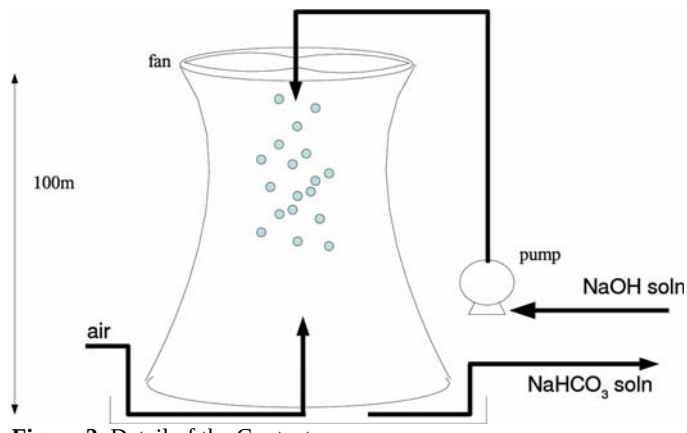

Figure 2: Detail of the Contactor

(Fukunaka, 1992; Spector, 1946; Greenwood, 1953), but the energy and material requirements are not known.

Methods

Numerical model. A numerical model was constructed of a single falling droplet of NaOH solution. The droplet falls by gravity through 100 *m* of air with a uniform velocity of 1 *m/s*. Mass transfer of CO_2 into the drop was modeled with boundary layer theory. Resistance in the liquid layer was considered to be negligible because internal circulation in drops of the size range considered is relatively fast (Pruppacher, 1978). Mass transfer through the air boundary layer was estimated with an empirical relation developed for raindrops (Bird, et al, 1960) and integrated over the fall of the drop.

Experimental. To confirm the theoretical calculations, experimental measurements of CO_2 uptake are performed. Droplets of 1*M* NaOH solution are ejected from an electrostatically-charged nozzle fed by a syringe pump. Droplet diameters between 0.3 and 1.5 *mm* are achieved with voltages of 3-6 *kV*. Droplets fall through a measured distance of atmospheric air and are collected in a buffered (pH~6) solution so that subsequent CO_2 uptake is negligible. The total carbonate in the buffered solution is measured using a model 1100 Total Organic Carbon (TOC) analyzer (OI Analytical). The analyzer acidifies the sample to pH<2 to convert all inorganic carbon to CO_2, purges the sample with N_2, and measures the resulting CO_2 with a nondispersive infrared detector (NDIR). Uptake of CO_2 per drop is calculated based on total CO_2 captured and the total volume

of solution collected. By repeating this experiment for a range of drop sizes an empirical prediction of CO_2 uptake as a function of drop size is generated.

Results

Results of the numerical model are shown in Figure 3. Concentrations of captured CO_2 that approach or exceed 1 *M* are predicted for drops smaller than about 1 *mm*. A more meaningful measure of uptake efficiency may be on a per-energy basis. Energy is required to pump the solution to the top of the tower and to sustain a large enough pressure head to break the solution into mist. The energy required to move air through the contactor was estimated to

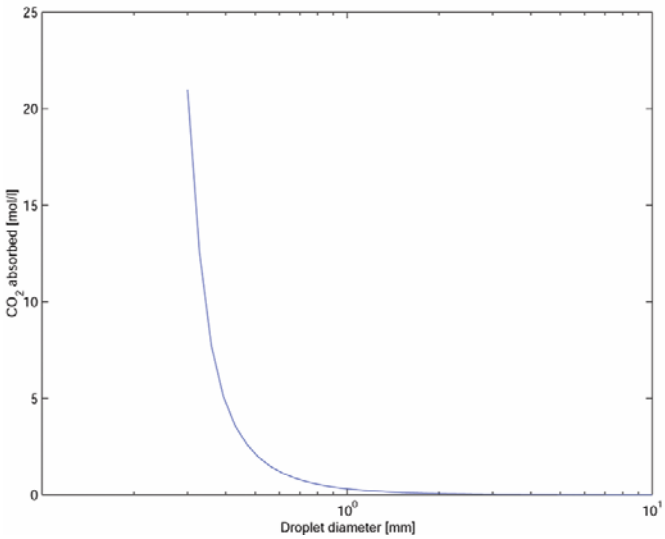

Figure 3: CO_2 absorbed by falling droplets as a function of droplet diameter

be small in comparison. Figure 4 shows the results converted to energy terms. For drop sizes smaller than 1 *mm* or so, the energy requirements are quite modest – less than 1 *kJ/mol* CO_2 for the smallest drops, which compares favorably with, for instance, the heat of combustion of gasoline at 680 *kJ/mol* CO_2. This is also small compared to our preliminary estimates of the energy requirements of the total system (primarily the calciner), which may amount to 220 *kJ/mol* CO_2.

Results of the laboratory experiments are pending.

Discussion

CO_2 capture from ambient air is an understudied but potentially important subset of CCS technology, having several strong advantages over CCS from point sources. The aim of this research is to help develop and analyze such a system that is simple, scalable, and achievable with current technology. The primary goal of this research is to develop a high-quality estimate of the cost of operation of the contacting unit, in dollars per ton of CO_2 captured, that can be used in the cost-accounting of the entire system. The insight and data gained from the prototype experiments, along with data and knowledge from industry contacts, will enable this estimation.

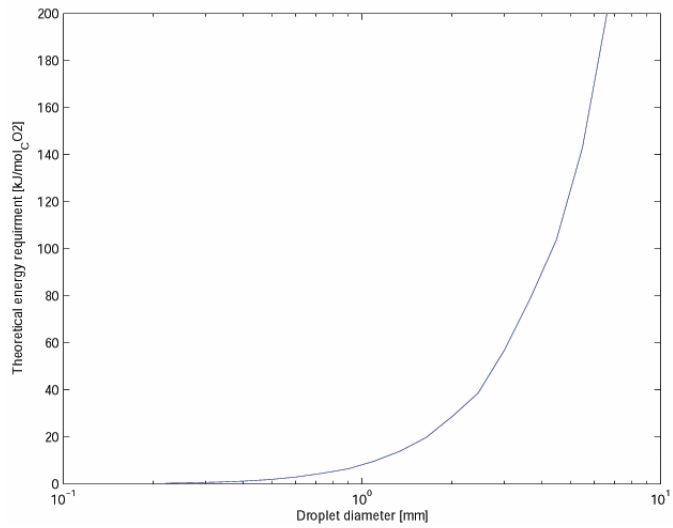

Figure 4: Theoretical energy required for CO_2 capture as a function of droplet diameter

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