

## Enhanced Practical Photosynthetic CO<sub>2</sub> Mitigation

D. J. Bayless ([bayless@ohio.edu](mailto:bayless@ohio.edu); 740-593-0264)

G. G. Kremer ([kremer@ohio.edu](mailto:kremer@ohio.edu); 740-593-1561)

Ohio Coal Research Center  
Department of Mechanical Engineering  
Ohio University  
251 Stocker Center  
Athens, OH 45701-2979

M. E. Prudich ([prudich@ohio.edu](mailto:prudich@ohio.edu); 740-593-1501)

B. J. Stuart ([stuart@ohio.edu](mailto:stuart@ohio.edu); 740-593-9455)

Ohio Coal Research Center  
Department of Chemical Engineering  
Ohio University  
172 Stocker Center  
Athens, OH 45701-2979

M. L. Vis-Chiasson ([vis-chia@ohio.edu](mailto:vis-chia@ohio.edu); 740-593-1134)

Environmental & Plant Biology  
Ohio University  
400 Porter Hall  
Athens, OH 45701-2979

K. Cooksey ([umbkc@gemini.oscs.montana.edu](mailto:umbkc@gemini.oscs.montana.edu))

Department of Microbiology, LW-113B  
Montana State University  
Bozeman, MT 59717-0326

J. Muhs ([um4@ornl.gov](mailto:um4@ornl.gov); 865-574-9328 *voice*; 865-576-0279 *fax*)

Oak Ridge National Laboratory  
P.O. Box 2009, MS-8058  
Oak Ridge, TN 32831

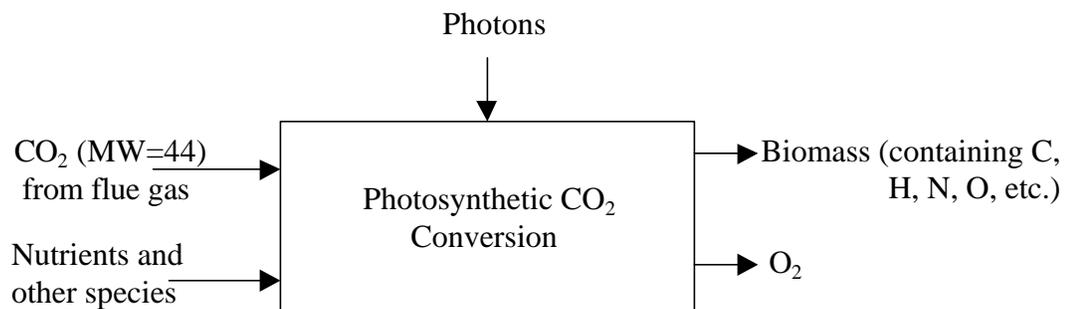
### INTRODUCTION

Biological carbon sequestration, in particular engineered photosynthesis systems, offers advantages as a viable near-to-intermediate term solution for reduced carbon emissions in the energy sector. Such systems could provide a viable option for “other-than-ocean” sequestration for smaller fossil generation units located in the midwest. Photosynthetic (or “natural” sequestration) systems produce usable byproducts (biomass). Further, such systems could minimize capital and operating costs, complexity, and energy required to transport CO<sub>2</sub> that challenge sequestration in deep aquifers or mines. Lower capital costs are extremely important, especially to small generators, who may not be able to afford separation and CO<sub>2</sub> delivery

systems that are only cost effective if done on very large scales. For coal to remain competitive, especially in the rapidly emerging distributed generation market (< 50 Mw), and ensure future fuel diversification, a portfolio of viable and practical sequestration techniques will have to be developed. Photosynthetic systems should be a part of that portfolio.

Despite the large body of research in the area of photosynthesis for carbon sequestration, little work has been done to create a practical system, one that could be used with both new and existing fossil generating units. For example, use of raceway cultivators ignores land availability limitations at existing fossil generation plants. Few existing smaller generation units could find 200+ acres of suitable land for siting a microbial pond much less build and maintain one throughout a midwestern winter. Additionally, how would the CO<sub>2</sub> be introduced to the photosynthetic agents? Would such a system need expensively separated CO<sub>2</sub> (not direct flue gas) for sparging? Would local stack emissions restriction prevent dispersion of flue gas at ground level? In addition, questions exist about supply and distribution of light. For example, in a raceway cultivator or pond, only organisms near the surface would receive sufficient photons for photosynthesis due to the high degree of reflection and attenuation caused by the water. If organisms had to exist at the surface (and outside), would cold weather have a negative impact on their performance? Further, to keep any such system operating at maximum carbon uptake rate, mature and dead organisms would need to be harvested. How would that be accomplished and at what rate? Finally, although numerous post-harvesting uses exist, what would be the optimal use with respect to the specific application and host site? These questions must be addressed before a practical photosynthetic system can be used.

The concept behind engineered photosynthesis systems is straightforward. Even though CO<sub>2</sub> is a fairly stable molecule, it is the basis for the formation of complex sugars (food) by green plants through photosynthesis. The relatively high content of CO<sub>2</sub> in flue gas (approximately 14% compared to the 350 ppm in ambient air) has been shown to significantly increase growth rates of certain species of microalgae. Therefore, application is ideal for contained systems, engineered to use specially selected (but currently existing) strains of microalgae to maximize CO<sub>2</sub> conversion to biomass, absorbing greenhouse gases. In this case, the microalgal biomass represents a natural sink for carbon.



**Figure 1.** Photosynthetic conversion of CO<sub>2</sub> to biomass and oxygen

As shown in Figure 1, if the composition of "typical" microalgae (normalized with respect to carbon) is CH<sub>1.8</sub>N<sub>0.17</sub>O<sub>0.56</sub>, then one mole of CO<sub>2</sub> is required for the growth of one mole of

microalgae. Based on the relative molar weights, the carbon from 1 kg of CO<sub>2</sub> could produce increased microalgal mass of 25/44 kg, with 32/44 kg of O<sub>2</sub> released in the process, assuming O<sub>2</sub> is released in a one-to-one molar ratio with CO<sub>2</sub>. Therefore, a photosynthetic system provides critical oxygen renewal along with the recycling of carbon into potentially beneficial biomass.

Enhanced natural sinks could be among the most economically competitive and environmentally safe carbon sequestration options because they do not require pure CO<sub>2</sub> and they do not incur the costs of separation, capture, and compression of CO<sub>2</sub> gas. Among the options for enhanced natural sinks, the use of existing organisms in an optimal way in an engineered photosynthesis system is lower risk, lower cost, and benign to the environment. This contrasts the use of ocean-based sinks, which could present problems. Large amounts of iron must be added to the ocean to promote additional CO<sub>2</sub> fixation. As a result, there may be little control over resulting growth. “Weed” plankton, the most likely organisms to grow, would not provide sufficient nutrients for the food webs, generating a high probability of negative environmental impact.

An engineered photosynthesis system could be placed at the source of the emissions to allow measurement and verification of the system effects, rather than being far removed from the emissions source, as is the case with forest-based and ocean-based natural sinks. The byproducts of an engineered system, biomass, could be used as a fuel, fertilizer, feedstock, or source of hydrogen. And even though some carbon is released from biomass through decomposition, bioconversion is the fastest and safest method to add carbon to natural terrestrial sinks. Further, the process described in this paper also requires relatively small amounts of space (an estimated 1/25<sup>th</sup> of a comparable raceway cultivator design). Because the organisms are grown on membrane substrates arranged much like plates in an electrostatic precipitator, there is little pressure drop. The system described here could be used at virtually any power plant with the incorporation of translating slug flow technology to create favorable conditions, such as reduced temperatures and enhanced bicarbonate concentration. Finally, engineered photosynthesis systems will likely benefit from current research into enhancing the process of photosynthesis, either genetically or via catalysts. This synergistic effect could lead to significant CO<sub>2</sub> reductions not otherwise possible.

Additionally, most of the required energy, even for auxiliaries, could be provided by collected sunlight. From a solar energy utilization standpoint, this work offers a unique and cost-effective alternative using a new hybrid system that leverages two decades of advancements and cost improvements in the solar, optical coating, and large-core optical fiber industries. This method far surpasses previous attempts at distributing sunlight to enhance microalgal growth.

## **OBJECTIVES**

The objectives of the effort described in this paper focus on the development of a practical photosynthetic system for greenhouse gas control with special focus on application at smaller fossil generation units. The work described here has focused on selection and study of viable thermophilic organisms, design of the growth surfaces within the bioreactor to reduce overall system size, photon collection and delivery via fiber optics to optimize growth and reduce system footprint, and harvesting schemes to facilitate maximum growth rates. Research has also

been directed to the application of translating slug flow technology to enhance concentrations of soluble carbon species to increase organism growth rates, which is also used to reduce flue gas temperatures. The ultimate goal is to test a complete pilot scale system to demonstrate process viability.

## PROJECT DESCRIPTION

The conceptualized process, shown schematically in Figure 2, begins after the flue gas has passed through suitable particulate control device(s) so that the gas will be substantially free of solid impurities. Then the flue gas must be cooled. Translating slug flow is used for both cooling the flue gas and generating soluble carbon species to “feed” the bioreactor. The water used in the process must also be cooled (using a cooling tower) due to solubility limitations of the water. The cooled flue gas, and separately the soluble carbon from the slug flow reactor, pass through the bioreactor, which houses vertically suspended growth membranes growing thermophilic organisms, arranged to minimize pressure drop of the flue gas throughout the reactor. The growth substrate must be resistant to wear in the harsh environment of the flue gas and corrosive potential of the growth media and, because of the vertical position, offer a high degree of adhesion with the microalgae. However, the degree of adhesion can be too high, becoming problematic for harvesting.

Solar photons are the energy source of the system and one of the primary factors determining system efficiency. In order to utilize solar photons at maximum efficiency, the system requires a light delivery subsystem to deliver a sufficient quantity and quality of photosynthetic photons deep within the bioreactor and minimize the light loss due to reflection and adsorption. Direct, filtered sunlight is collected and delivered into the bioreactor, via collection optics and large-core optical fibers. Visible light from the sun reflected from 1.5-meter diameter dish collectors and secondary optics is launched into approximately thirty-five 15 mm optical fibers (seven rows of five fibers). Large core fiber optic cables then supply photons necessary to support photosynthesis, using special distributors located between the vertical growth membranes. By controlling attenuation through the fiber optic cables, a uniform distribution of photons, typically at a rate between 40-80  $\mu\text{mols m}^{-2} \text{s}^{-1}$ , is supplied. This distribution is a key element in reactor design. The sunlight, originally collected by tracking mirrors (optimizing solar collection) will provide over 2000  $\mu\text{mols m}^{-2} \text{s}^{-1}$  of suitable photons throughout the day. However, at that rate, most photons would be wasted, as photosynthesis in thermophiles occurs at much lower levels of light. Therefore, distribution and diffusion of the photons is critical to bioreactor operation. A further point of interest is that sunlight contains wide spectra of energy; some is useless to the photosynthetic organisms, such as infrared, and some is harmful, such as certain ultraviolet spectra. Filters on the entrance to the fiber optic cables remove unwanted portions of the solar spectra and allow it to be used for photovoltaic production of electricity needed to power the auxiliary components of the system.



The harvesting system provides a way to remove mature organisms and repopulate the membranes with developing organisms, thus maximizing carbon uptake. Preliminary tests indicate that microalgae, removed in "clumps" from the growth strata, are easily agitated into a diffuse state. Mature or dead microalgae (organisms with a low potential for carbon utilization) can be removed and microalgae that are maturing, (organisms with a high potential for carbon utilization), can be repopulated on the growth strata. The harvesting process is also necessary to promote cell division and to reap the benefits of post-processed biomass.

One of the more significant engineering challenges of this project is nutrient enhancement and delivery to the photobioreactor. Microalgae often more easily fix carbon and nitrogen in soluble form. Translating slug flow technology, developed at Ohio University's Institute for Corrosion and Multiphase Processes, not only increases concentrations of nutrients in the aqueous phase by directly removing them from the flue gas, but also lowers flue gas temperatures. Slugs create zones of greatly enhanced gas-liquid mass transfer, putting CO<sub>2</sub> and NO<sub>x</sub> into soluble form for the microalgae. Significant work remains to optimize the nutrients to maximize photosynthetic growth and to maximize the transport capability of water with respect to solubility.

Perhaps the single most important factor that could result from translating slug flow use could be elimination of direct exposure of the photosynthetic organism in the bioreactor to flue gas. If the water from the slug flow reactor contained more than enough soluble carbon species and other nutrients to feed the organisms, further exposure to flue gas might become pointless. This would offer the advantage of using less thermotolerant microalgae, promoting even greater carbon utilization rates, because the water temperature from the slug flow reactor would be between 35-40°C (as opposed to 60°C for flue gas), and result in possible cost savings.

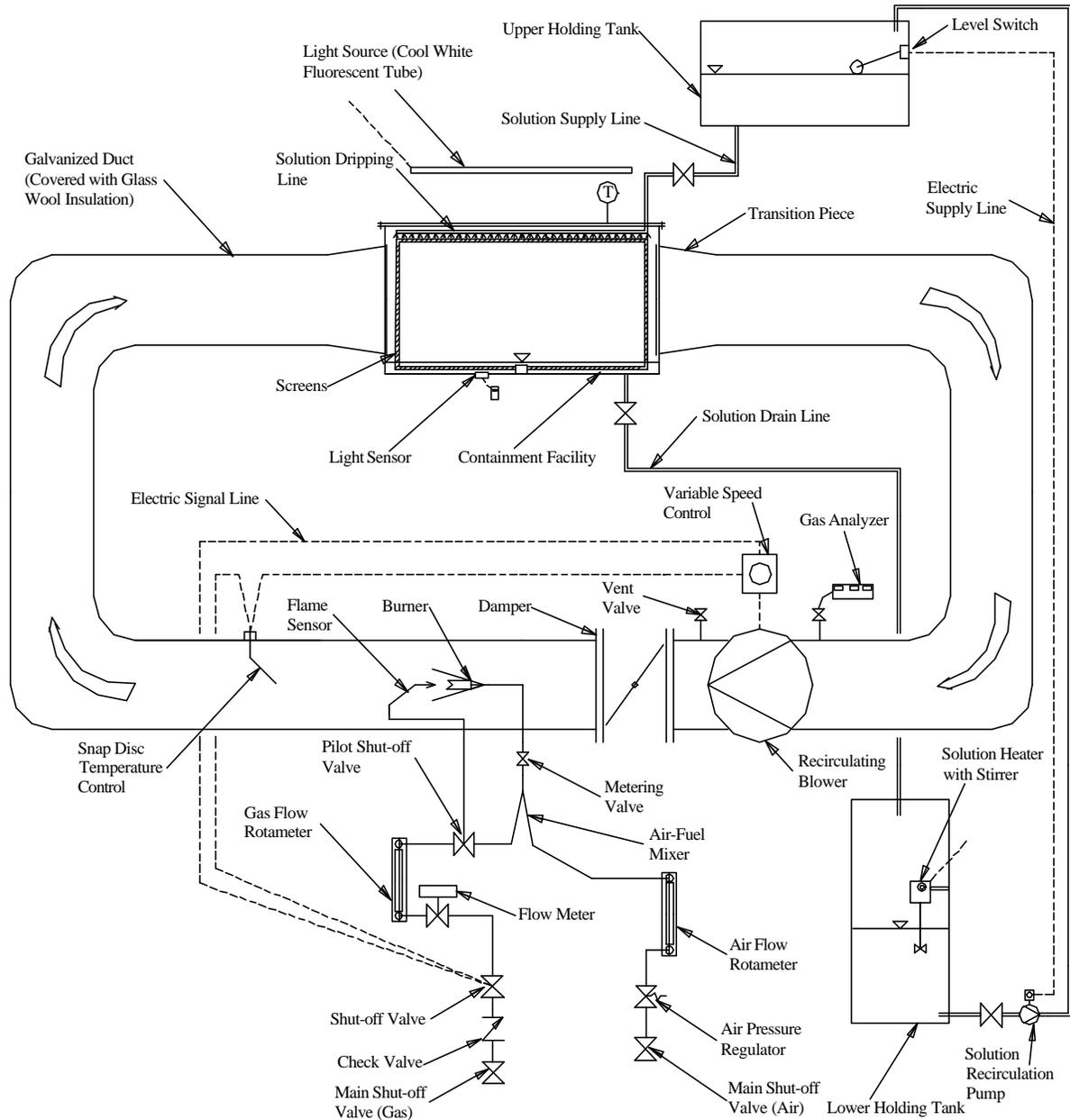
### **APPROACH (Methodology)**

In order to test our photobioreactor design, the Carbon Recycling Facility (CRF), shown schematically in Figure 3, was constructed. The CRF is a bench-scale test facility designed to simulate the flue gas emission from fossil-fired power plants. It has four subsystems in addition to the bioreactor: the flue gas circulation system, the gas burner system, the algal growth solution circulation system, and the analysis system.

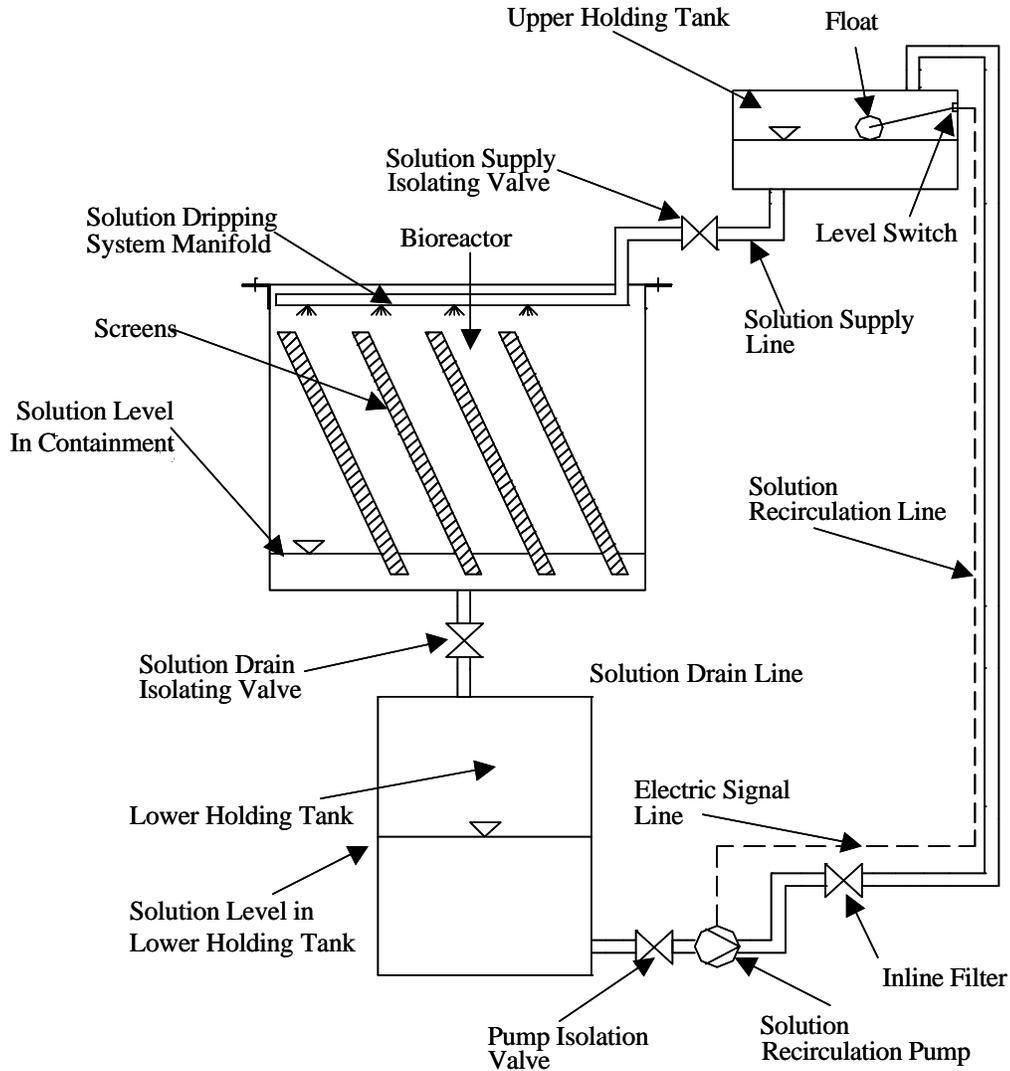
The flue gas circulation system was designed to recirculate hot gas, rich in CO<sub>2</sub>, through the bioreactor by using a contained fan. The gas burner system was designed to maintain a suitable operating temperature for the circulating gas, while providing sufficient levels of CO<sub>2</sub>. The burner assembly consists of a burner nozzle, covered with a galvanized cone so as to protect the root of flame from the flow of circulating gas. The sub-assembly of burner nozzle, cone and flame sensor was mounted inside the ductwork and serves as a combustion chamber. Burning natural gas, which is free from any solid impurities, creates the simulated flue gas. Temperatures may range from 40°-70° C and CO<sub>2</sub> concentration can vary up to 20% (v/v).

The solution circulation system was designed to provide culturing media to the algal culture dispersed over vertically suspended screens while they are subjected to the high temperature

simulated flue gas. This system keeps the algae moist inside the bioreactor and provides nourishment. The system is illustrated in Figure 4.



**Figure 3.** Schematic flow diagram of Carbon Recycling Facility



**Figure 4.** Schematic flow diagram showing growth solution recirculation system.

The containment facility, also known as the bioreactor, is a box made of 1/4" thick plexiglass, with top cover made of 3/8" thick plexiglass. Plexiglass was chosen because it can sustain high temperatures and is transparent. Four screens made of polyester fabric cloth fastened within the frame and loaded with algal culture are placed inside the containment at an angle of 85° from horizontal. Screens are 21" long x 10½" wide with ½" frame width. The growth solution is dripped over the screens from an upper holding tank made of opaque plastic with a capacity of 18 gallons, through a manifold which serves to drip solution. Solution collected in the containment is drained back to the lower 24 gallon holding tank made of steel. Both tanks are black so as to prevent any photosynthetic reactions. Algal solution from the lower holding tank is pumped back to the upper holding tank by a circulation pump. An inline 0.5 µm rated filter is used to trap algae.

Flue gas is continuously analyzed for CO, CO<sub>2</sub> and O<sub>2</sub> content using a Nova Analytical Systems Inc., model 375WP analyzer. The analyzer utilizes a sensitive infrared detector for CO<sub>2</sub> and disposable electrochemical sensors for O<sub>2</sub> and CO. The temperature of the flue gas is analyzed by inserting the thermocouple wire into the flue gas stream in the containment. The pH of the circulating solution is measured using Hanna Instruments pH meter, model pHep. The range for the pH meter is 0.0 to 14.0 pH with resolution of 0.1 pH and accuracy (@20°C) of ±0.1pH. Photosynthetically Active Radiation (PAR) is measured using a Licor LI-190SA quantum sensor. The quantum sensor measures PAR received on a plane surface. A silicon photodiode with an enhanced response in the visible spectra is used as the sensor.

For each experiment, the steps involved were: preparing algal culture, sampling algal culture to determine mass of algae used for experimentation, preheating of the containment facility and culturing solution, setup for the analysis instrument. To begin, the microalgae were cultured in 20 gallons of growth solution designated as medium I. The culture was illuminated by a 42W cool-white fluorescent lamp at an intensity of 64 μmol·s<sup>-1</sup>·m<sup>-2</sup> and bubbled with air and CO<sub>2</sub> mixed together in the ratio of 19:1. The fluidization created by the bubbled CO<sub>2</sub> helped in uniform defragmentation and CO<sub>2</sub> transport via bulk flow.

The cultured algal colonies were transferred into a 6-gallon plastic sampling containment, from where the algal samples are drawn out for experimentation. The containment was provided with a sample draw out isolation valve and a closed circuit heater to maintain the algal samples at 45° C and pH level of 7.4 to prevent thermal shock upon entering the bioreactor.

After being cultured and pre-heated, the samples were withdrawn. Each sample was 25ml by volume and filtered through pre-weighed filters. The filters were heated in an oven at 65°C. Dry mass of algae in each sample was determined from the difference between the clean, unused filter and the algae-containing filter. A statistical analysis of the dry weights provided the dry weight concentration of the algae in the overall sample.

For each experimental run, the containment facility and algal growth solution were preheated for 12 hours to stabilize at the specified temperature. Additionally, between each experimental run, the containment facility and the upper and lower holding tanks were cleaned and a new pre-weighed dried element was installed in the inline filter.

## **RESULTS**

Experiments were performed to examine the effects of temperature, CO<sub>2</sub> concentration, and light intensity on the growth characteristics of microalgae species Nostoc 86-3 in our bioreactor. Healthiness and growth of algal samples were determined by visible inspection of color in the organisms, cellular structure and density, and difference in sum of dry weight of screens and inline filter cartridge when compared to the amount of algal sample loaded.

### ***Effect Of Temperature***

Two sets of experiments were conducted to study the ability of microalgae species to sustain a temperature of 150°F. These experiments were carried out for 150 hours with the experimental

containment illuminated by a bank of 60W cool-white fluorescent lights of  $33 \mu\text{mol}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$  intensity measured over the top cover of the bioreactor. It was observed from these experiments that cyanobacteria samples were green for approximately 100 hours and then changed in color to brown. The density of cyanobacteria samples over the screens was also drastically reduced. The results indicated that the cyanobacteria were unable to handle either the high temperature, the thermal shock resulting from the transition from room temperature, or both.

The next experiments were carried out at 130°F. Experiment III was carried out for 120 hours under an illumination of  $33 \mu\text{mol}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$  measured at the top cover of the bioreactor. Again, it was observed that the algal density decreased, indicating that there was no significant growth. The microalgae also showed change in color from green to brown after 96 hours.

Because of poor growth performance at 130°F, the next experiment was carried out at 120°F with an average photon intensity of  $33 \mu\text{mol}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$  measured at the top cover of the bioreactor. The simulated flue gas composition at 120°F was identical to one for Experiment III. After the experiment was run for 120 hours, the growth screens and filter were removed and dried. The test results infer that the differential weights of algal samples after the trial were less than the original loaded weight and that cells showed a change in color from green to brown after 110 hours. However, microscopic analysis revealed that the samples were healthy with cells still forming chains although the average cell size was 50% of the original culture.

### *Effects of Light Intensity*

The next experiment was carried out at 120°F, which was established as the upper limit for this species, but with a higher luminance of  $55 \mu\text{mol}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$  measured at the base of the experimental containment, after the growth samples were loaded over the screens. Note that the light was constantly applied to the bioreactor. Again it was found that cells changed from green to brown within 80 hours due to the cells developing a protective pigment as a response to the high light intensity.

The next experiment was again performed at 120°F, but at a light intensity of  $18 \mu\text{mol}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$  measured at the base of the bioreactor after the algae were loaded over the screens. It was observed during the experiment that the organisms remained green, but with reduced density on the screens. In addition, it was observed that amount of light intensity passing through the containment showed a continuous rise with time, indicating a loss of cell density. However, the mass obtained after the trial was more than that initially loaded, indicating positive growth.

The next experiment was conducted at 120°F at an illumination of  $22 \mu\text{mol}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$  measured at the base of the bioreactor after the algal samples were loaded. In this experiment, instead of applying continuous lighting, the organisms were exposed to a 12-hour lighting cycle. The results showed a dramatic improvement in growth. Further, it was observed that the light intensity passing through the containment showed only a small increase. It was also observed that the cells showed a slight change in color. However, cellular studies indicated that the species were of consistent size with the batch culture and maintained the filamentous morphology of *Nostoc*. Based on visual observations, the species appeared viable.

These results were augmented by measurements of CO<sub>2</sub> loss from the bioreactor test section during this test. The corresponding mass balance was resolved to within 2%, which is remarkable for the low level of CO<sub>2</sub> actually absorbed. The net result was approximately 10.2 g of CO<sub>2</sub> absorbed of the original 2.97 m<sup>3</sup> of circulating flue gas, (or about 19% of the original CO<sub>2</sub>). While this result in no way predicts the ability of the system to remove CO<sub>2</sub> over the long term in a full-scale operating system, it appears to give credence to the workability of the system.

The final tests were done at more uniform light intensities (with 12 hour lighting cycles), with ~ 40 μmols-s<sup>-1</sup>m<sup>-2</sup> at the base of the bioreactor. The results using a more uniform light distribution at 10, 15 and 20% CO<sub>2</sub> levels all indicate positive growth (with an average 30% net mass gain after 120 hours).

## **BENEFITS**

Four major benefits, in addition to CO<sub>2</sub> mitigation, could result from the use of this novel method of photosynthetic sequestration. These benefits are electrical power generation (from photovoltaics), oxygen production, reduction of gaseous pollutants including potential NH<sub>3</sub> slip (from selective catalytic reduction to control NO<sub>x</sub>) and NO<sub>x</sub>, and production of biomass with beneficial end-uses. Oxygen is a natural product of photosynthesis. If you assume that 1 mole of O<sub>2</sub> is formed for each mole of CO<sub>2</sub> consumed during photosynthesis, then for every kg of CO<sub>2</sub> consumed, (32/44) or 0.73 kg of O<sub>2</sub> are produced. While this may not seem terribly significant, one can not overestimate the importance of oxygen to our lives. In terms of other pollution control, this process could provide NO<sub>x</sub> control at no additional cost. First, the translating slug flow process used to enhance soluble carbon concentration is a natural scrubber. Not only is NO<sub>x</sub> converted to nitrates, SO<sub>x</sub> is converted to sulfates and sulfites, and any NH<sub>3</sub> that might ‘slip’ through an upstream SCR process for NO<sub>x</sub> reduction will be scrubbed as well. Both NO<sub>x</sub> and NH<sub>3</sub> scrubbing are not only an additional benefit, such scrubbing is beneficial to photosynthesis, as the microalgae require nitrogen to grow. In fact, work by Yoshihara et al. (1996) shows considerable nitrogen fixation from NO<sub>x</sub> species bubbled through a bioreactor with poor mass transfer characteristics.

Finally, it should be noted that the resulting biomass has numerous beneficial uses. In addition to being a potential fuel, microalgae have been used as soil stabilizers, fertilizers, in the generation of biofuels, such as biodiesel and ethanol, and to produce H<sub>2</sub> for fuel cells. In recent tests, it also has shown suitable ignition characteristics to be cofired with coal in pulverized coal-fired generation units.

### ***Expected cost of commercializing in dollars per ton of carbon emission avoided***

Assuming a plant lifetime of 30 years and an 8.8% auxiliary load for pumping and dewatering (which would be lowered to 2.8% when the photovoltaics were operational) at an average cost of \$0.035 per kW-hr, a labor cost of \$1 per ton (mostly for hauling the dry biomass) and a comparable production price of a similar sized ESP (scaled by a factor of five (5) for the solar collectors), yields an approximate cost of \$8-\$10 per ton of CO<sub>2</sub> removed. The breakdown costs,

per unit ton of CO<sub>2</sub> removed, are \$4.50 capital cost, \$2-3 for operating costs, and \$1-\$2 for associated operating labor.

The optimistic potential long-term cost includes only \$1.50 per ton for power consumption because of the high level of self-generated (photovoltaic) power. Also note that the overall capital cost used in this estimate was approximately 40% greater than the estimate independently provided by the lighting team at Oak Ridge. If their estimate is used, the cost per ton of CO<sub>2</sub> removed decreases another \$0.50. Finally, the calculations for cost also do not include any potential revenue from the sale or use of the biomass, which would further reduce overall costs or credit for generation of O<sub>2</sub>.

## **FUTURE ACTIVITIES**

The results of this study to-date indicate that further work is needed before such a system could be considered for practical application. Specifically, issues of light delivery and distribution, enhancing growth rate through increased bicarbonate concentration, flue gas cooling, and harvesting to provide sustained growth, must be addressed for long-term, full-scale functionality.

The investigation did reveal that potential solutions to these concerns exist. Regarding the issue of lighting, Oak Ridge National Laboratory's work on hybrid fiber-optic lighting offers great promise for application to this work. Not only is light collected more efficiently by tracking the sun, photosynthetic photons can be distributed more efficiently by reducing overall photon concentrations to levels more suited to sustainable organism growth. The problems of both soluble carbon concentration and flue gas cooling could be addressed by employing translating slug flow. Harvesting was only briefly investigated using a number of mechanical processes, with simple water jet spray cleaning providing the least damage to the organisms and consistently removing 30-40% of organisms from the membrane substrate. More work in these areas is necessary to develop a truly viable photosynthetic process.

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