DEVELOPMENT OF NOVEL ACTIVATED CARBON COMPOSITES

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INTRODUCTION

The development of a novel activated carbon composite, carbon fiber composite molecular sieve (CFCMS), at the Oak Ridge National Laboratory has been discussed in earlier reports. Here, we report the investigation of CO_2 capture and separation from feed gases of varying CO_2 concentrations using CFCMS. The effect of varying the property of the CFCMS, characterized by the percent of the material that was burned-off during its activation, on the capture and separation of CO_2 was investigated. Because of the high thermal and electrical conductivity of the CFCMS, a new process named "electrical swing adsorption" (ESA) was developed. In the ESA process, a large amount of the adsorbed CO_2 can be desorbed by the application of a low voltage across the adsorbent. Several regeneration processes were examined in order to demonstrate and understand the ESA process.

PROCEDURE

Cylindrical billets of CFCMS, ~4.5-inches in diameter by lengths varying form 7.5 to 10 inches depending how much material was formed and on the percentage of it that was burnt-off, were processed. The burn-off amount varied from 19.0 to 42.9% resulting in different void fractions, capacities and other properties. Each cell was prepared by machining the CFCMS billet down to a diameter of 4.25 inches so that electrodes can be attached to it to power the cell during regeneration. The design is such that the feed gas has a mixing chamber both at the entrance to and exit from the cylindrical cell. The electrodes and other required mountings were shrink-wrapped onto the cell to form an airtight system. The sketch of the CFCMS cell is shown in Fig. 1. The cell was then mounted into the modified gas flow loop shown in Fig. 2. Flow through the cell can be reversed. There is a by-pass that carries the feed gas directly to the sampling system, which consists of a real-time mass spectrometer, residual gas analyxer, and a real-time CO_2 analyzer.

The feed gas is passed through the cell and then to the sampling system. The flow rate in and out of the cell and the pressure drop across it are measured. The inlet gas temperature, the temperature, one inch into either end of the cell, and the temperature on the outside of the cell can all be recorded. The inlet gas pressure is usually set at 10 psig at the compressed cylinder and the gas was carried in 0.375-inch (9.5 mm) stainless steel piping about 20 feet (~6.1 m) to the entrance of the CFCMS cell. All the experiments were run at ambient temperature unless otherwise stated.

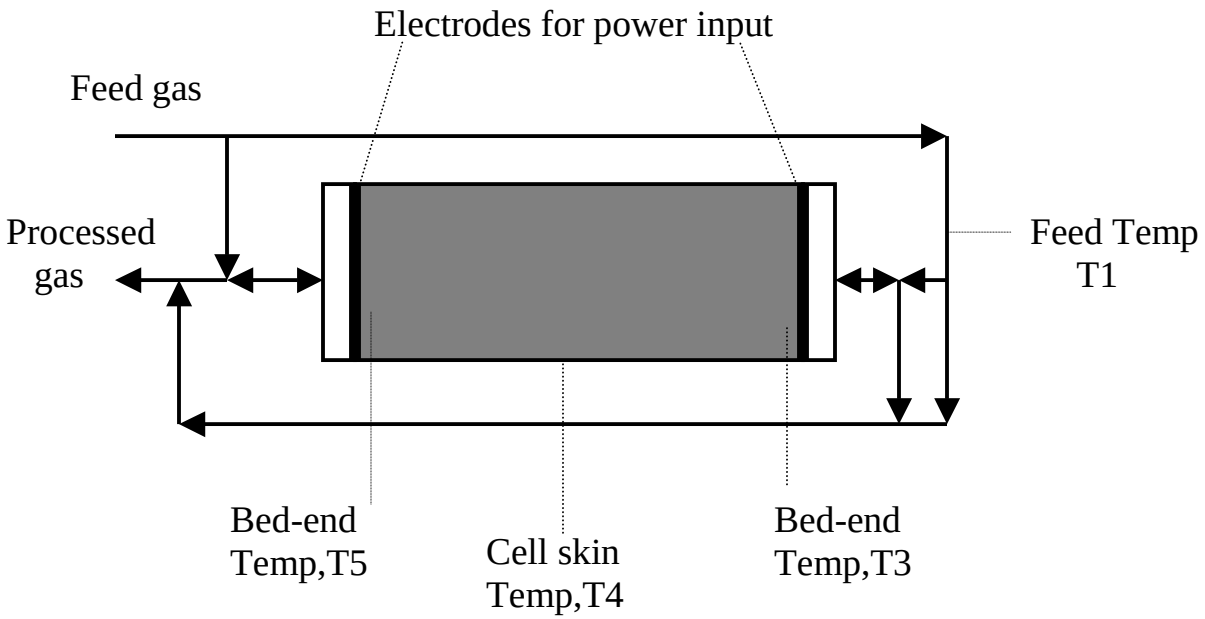


Fig. 1. The schematic of the CFCMS cell set-up.

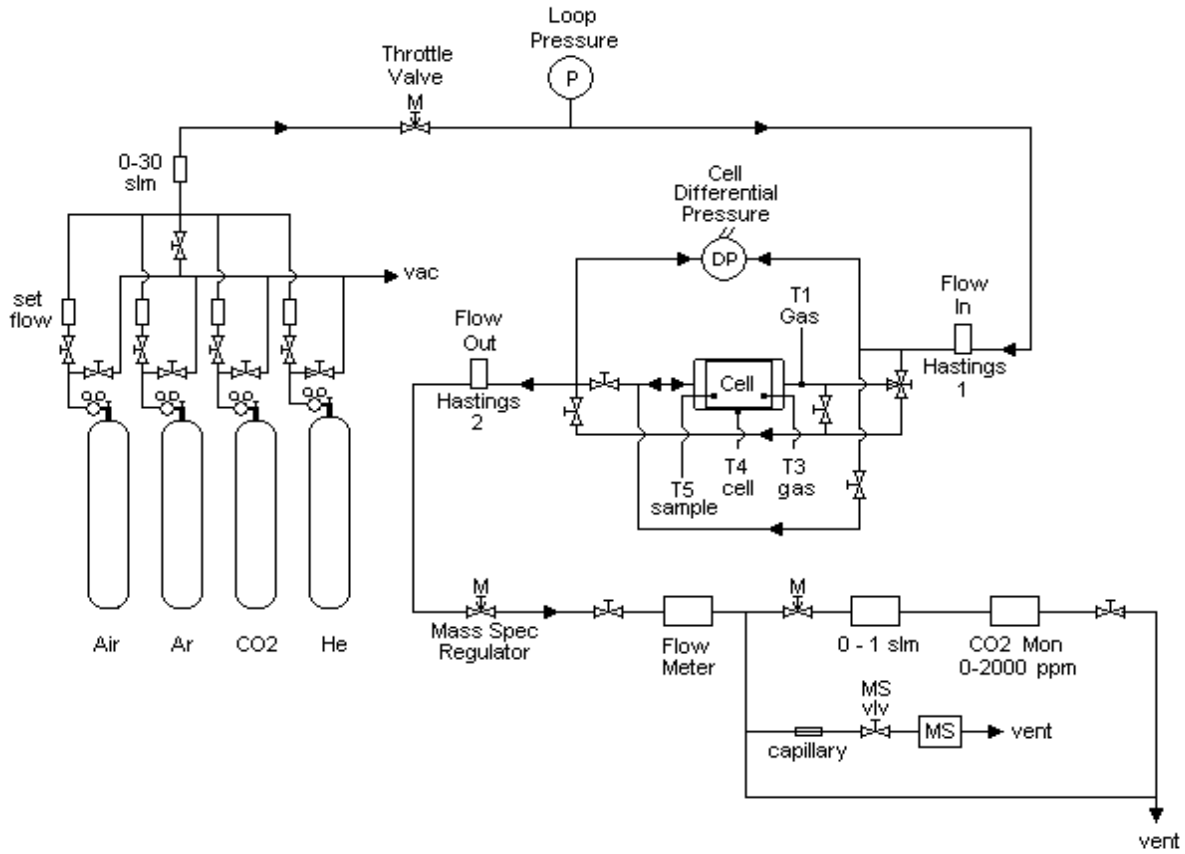
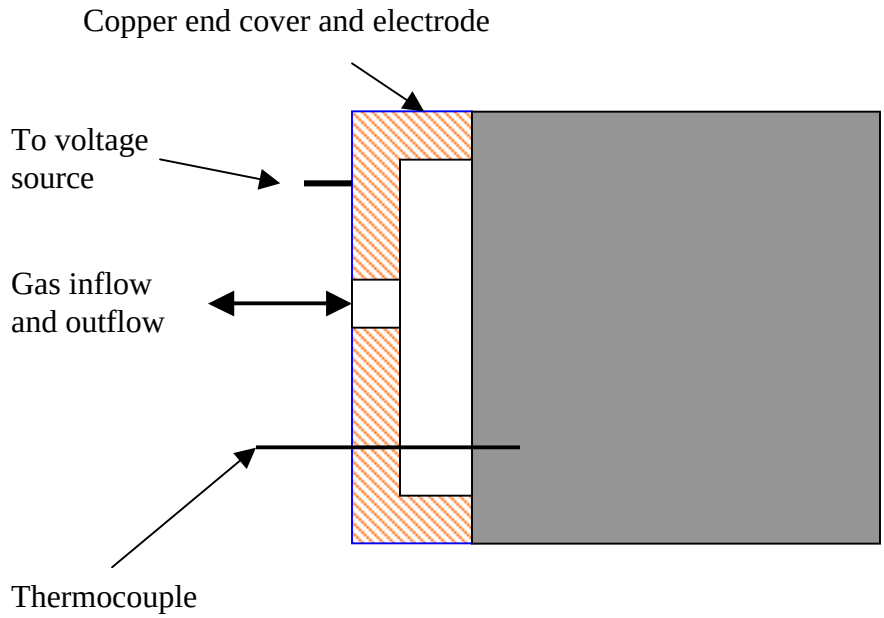
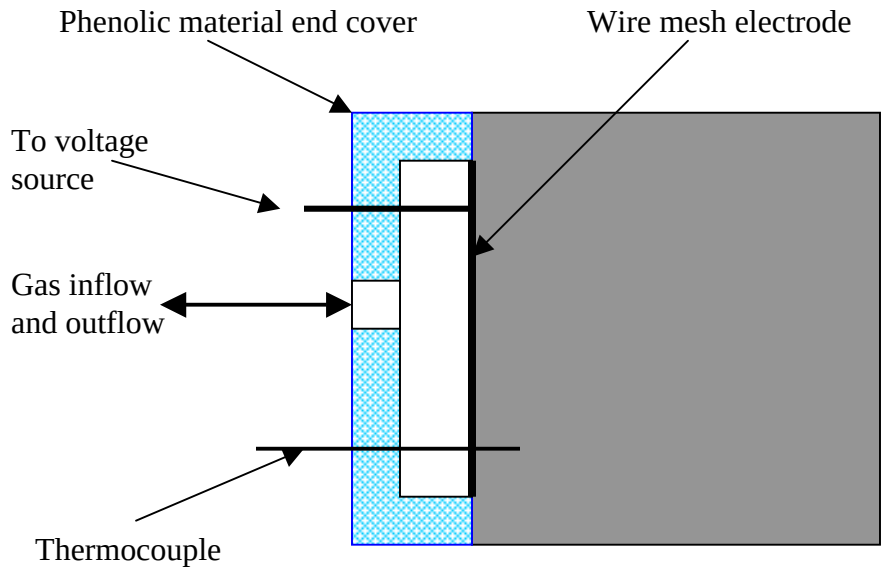


Fig. 2. The modified gas flow loop.

Two methods of attaching the electrode to the cell have been investigated. Their design is illustrated in Fig. 3(a) and 3(b), showing only an end of the cell. The first electrode was made of copper and the second was made of steel wire mesh.



(a)



(b)

Fig. 3, (a) Schematic of a cell end showing copper end cover as an electrode. (b) Schematic of a cell end showing phenolic end cover and wire mesh electrode.

To make the copper electrode, a piece of copper disc was machined as the end cover for the CFCMS billet and to provide for gas mixing at the entrance and exit of the cell. The edge of the copper forms a ring pressing on the CFCMS and this becomes the electrode through which power is supplied to the cell. This is shown in Fig. 3(a). The end cover is then shrink-wrapped over the CFCMS billet to form the cell.

Figure 3(b) shows the schematic for the wire mesh electrode. The end cover here is fabricated from a non-conducting phenolic material. The electrode is a disc cut from a stainless steel wire mesh. The mesh sits on the CFCMS material and is held on by the end cover. The end cover is shrink-wrapped over the CFCMS cell to form the cell. The wire mesh electrode design is preferred and is now used in all the experiments

Three parameters were set during each experiment: the feed composition, the feed flow rate, and the CFCMS percent burn-off. The CO_2 composition in the feed varied from ~400 ppm in compressed air to pure CO_2. The feed rates were from a fraction of a liter to 20 liters per minute (l/m) and the CFCMS burn-off varied from 19.0 to 42.9 %. CO_2 breakthrough curves were run at a set of these parameters and from them the amount of CO_2 adsorbed was calculated per gram or liter of the CFCMS adsorbent. The results of some of these experiments are reported below.

RESULTS AND DISCUSSIONS

Figures 4 and 5 show the breakthrough curves for a feed containing 10 mol% CO_2 flowing through a cell of 24.8% burn-off CFCMS at 2 l/m. Figure 4 is the breakthrough curve measured by the CO_2 monitor with a capacity limit of 20,000 ppm (2%). Figure 5 is the same breakthrough measured by the Residual Gas Analyzer and it shows all the other gases in the feed. Figure 4 indicates how the total gas scrubbed is calculated, and from this, the amount of CO_2 adsorbed is computed. Figure 4 also shows the temperature variation at the ends of the cell due to the heat of adsorption/desorption.

Similar breakthrough curves were run at 2 l/m for feed with varying compositions of CO_2: ~400 ppm (air), 3 mol %, 10 mol%, 19.5 mol% and pure CO_2 through cells with 24.8% and 37.8% burn-off CFCMS, respectively. The result is summarized in Fig. 6. The graph shows that whereas the specific adsorption of CO_2 by the CFCMS is dependent on the CO_2 feed composition, the relationship is not linear.

Breakthrough curves were run for a 10-mol% CO_2 feed at several flow rates varying from 1.0 to 20.0 l/m through cells containing CFCMS activated to four different % burn-off's. The results are summarized in Fig. 7. These results indicate that the dynamic capacity (g CO_2 adsorbed/g of CFCMS) of the CFCMS adsorbent is dependent on both the flow rate and the treatment (% burn-off) of the CFCMS. The results also indicate that optimum capacity in these runs was at feed flow rate of 10 l/m and for CFCMS activated between 25 and 35 % burn-off.

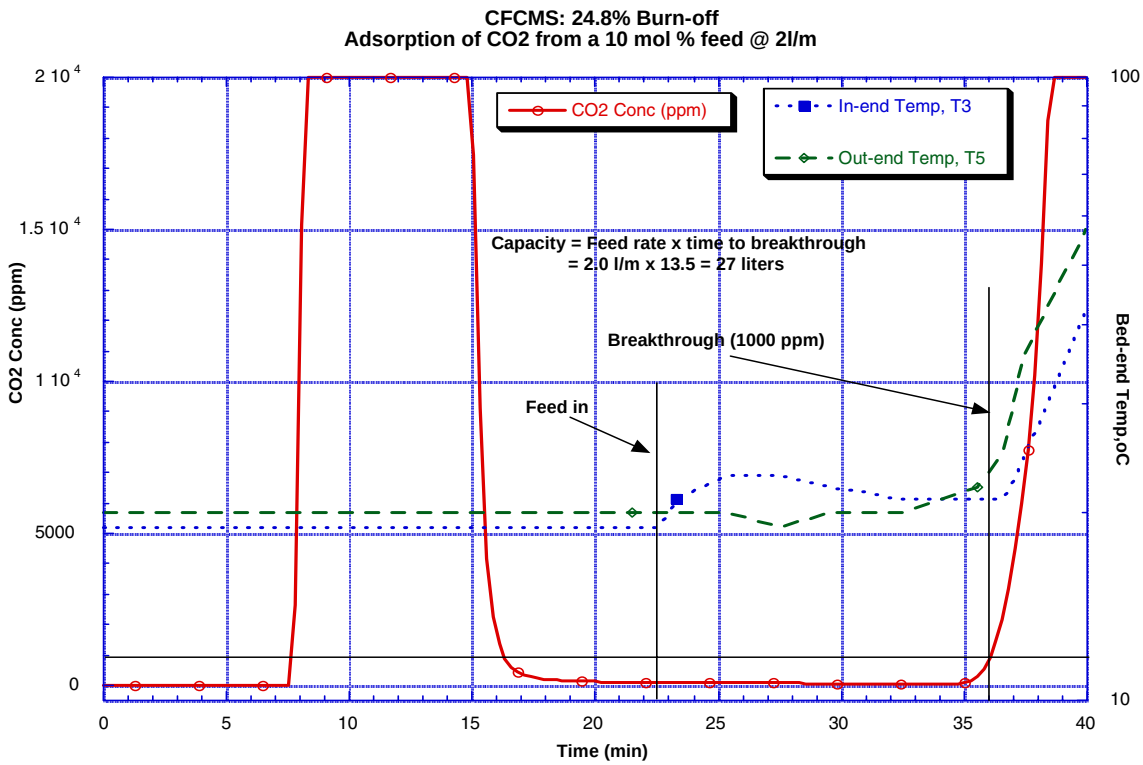


Fig. 4. Breakthrough curve on the CO_2 monitor for 10% CO_2 feed at 2 l/m.

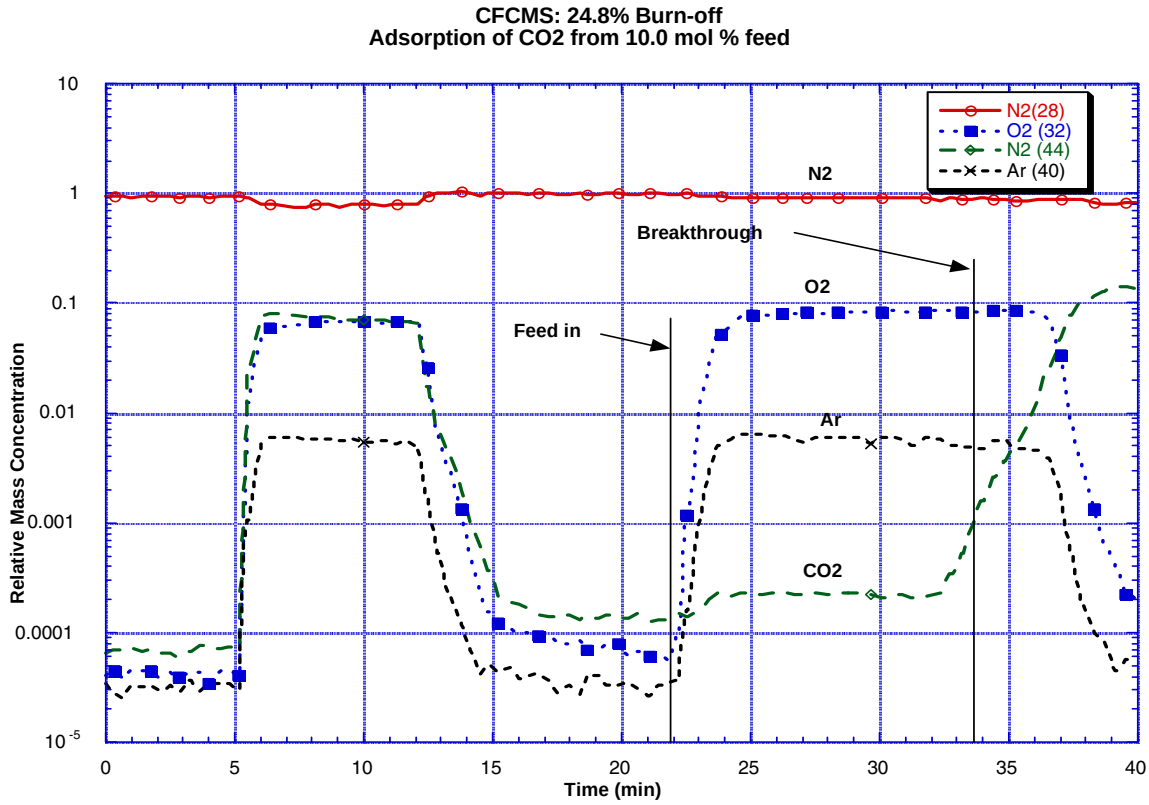


Fig. 5. Same breakthrough as in Fig. 4 but measured by the residual gas analyzer.

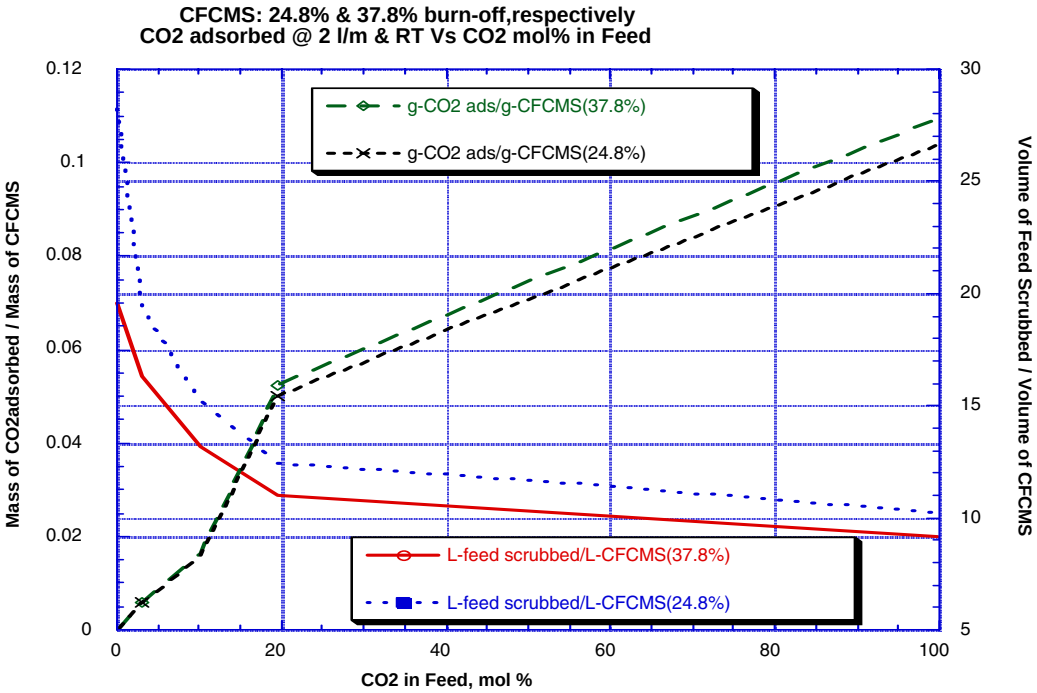


Fig. 6. Specific adsorption of CO_2 vs CO_2 mol% in the feed.

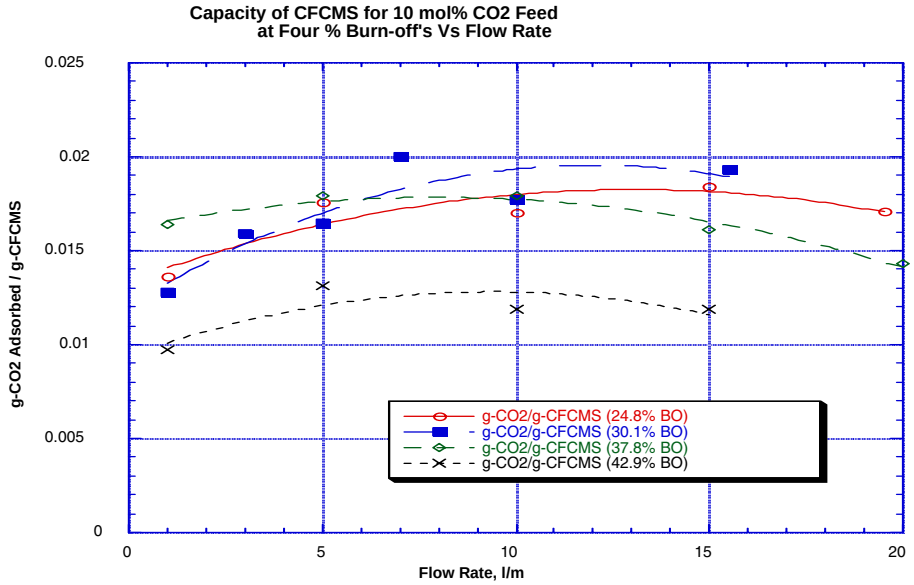


Fig. 7. Specific adsorption of CO_2 at four CFCMS treatments vs flow rate.

Pressure drop is an important parameter in determining the energy requirements for the flow through the cell. The pressure drop across the cell was measured during all the experiments. For all the four treatments (% burn-off) and flow rate 1–20 l/m, the pressure drop ranges from 0.003 to 0.02 psi/inch. The pressure drop is low because the CFCMS material is very porous with typical bulk density varying between 0.16 g/cc and 0.18 g/cc.

Regeneration is an essential step in using the CFCMS in a sorption process. Therefore, for a the CFCMS/ESA process to be viable, a well-defined regeneration step must be developed and optimized so that it consumes minimum energy and regenerant Consequently, several regeneration steps were investigated after the cell was saturated (CO_2 breakthrough at 350 ppm) at 5 l/m and feed flow stopped. N_2 flow at 1 l/m was used to complete the regeneration. The amount of N_2 required to complete the regeneration indicated the efficiency of the regeneration process.

1. **Temperature only.** Low voltage was passed through the cell until the temperature reached 70°C and was maintained there for several minutes. The flow through the cell was negligible. The N_2 needed for regeneration to complete the cycle indicated that the regeneration was very inefficient.
2. **Power only.** Low voltage was passed through the cell for 5 minutes as its temperature rose. No flow was detected. The regeneration was very inefficient.
3. **Temperature and Vacuum.** The cell temperature was raised to 70°C and maintained while the cell was put under full vacuum. There was some flow that resulted in some CO_2 removal. This process was more efficient that the first two.
4. **Vacuum only.** The cell was put under full vacuum for 5 minutes and its temperature fell. This process was more efficient than the first two but was worse than process 3.
5. **Power and Vacuum.** Low voltage was passed through the cell simultaneous with full vacuum on it. This was the most efficient regeneration process.

In all the processes, N_2 or an inert gas was needed to complete the regeneration cycle and prepare the cell for the next adsorption step. Furthermore, it seemed that only negligible pure CO_2 was produced in any of the processes. Consequently, the regeneration step requires further investigation in the CFCMS/ESA process.

CONCLUSIONS

1. Carbon fiber composite molecular sieve (CFCMS) adsorbs CO_2 completely from all feed gases until breakthrough occurs. The amount of feed gas scrubbed depends on the concentration of CO_2 in it.

2. The dynamic capacity of the CFCMS depends on both the treatment (%burn-off) of the CFCMS and the feed flow rate. The highest capacity was at feed flow rate of 10 l/m and CFCMS burn-off at 30 +/- 5%.
3. The pressure drop across the cell was very low varying between 0.003 and 0.02 psi/inch.
4. The most promising regeneration step is one that combines simultaneous power input to and vacuum on the CFCMS cell. However, further investigation is needed here to develop an efficient regeneration step that produces some pure CO_2.

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