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CYCLIC OPERATION OF ADSORPTION BEDS FOR USE IN SPACECRAFT LIFE SUPPORT SYSTEMS

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I. Objective

The objective of the investigation is to apply various cyclic modes of operation to the carbon dioxide recovery system of the Apollo spacecraft life support system in order to obtain improved performance. The cyclic mode has potential application to several other components of the H_2O and CO_2 recovery cycles.

II. Progress of the Investigation

1. Summary of previous investigations. It is generally believed that the optimum operating point of a chemical process system is the steady state. This viewpoint has recently been challenged, on the basis of evidence that nonlinear systems may achieve enhanced output when operated in unsteady fashion, i.e., cyclically.

This phenomenon has long been known in electronic technology as "parametric amplification," in which one field is enhanced at the expense of another, coupled field. In chemical processes, corresponding coupled fields would logically be sought in mass-transport and thermal-transport interactions. This has been proposed by Wilhelm and co-workers[1, 2], who refer to the effect as "parametric pumping" by analogy to the electronic terminology. Douglas and Rippin[3] have demonstrated that the output of a particular nonlinear reaction system is enhanced by cyclic operation, and several other investigators have reported studies in mass-transfer operations[4, 5, 6]. It is reliably reported that numerous industries and research groups are studying cyclic operation, in an effort to improve the output of various process systems.

Any process system that features coupled fields is potentially capable of enhanced operation through cycling. However, it is easily demonstrated that a linear uncoupled system, when subjected to a cyclic forcing function, will feature no enhancement of output. Therefore the systems of interest must be sought among those that feature transport fields that are (1) nonlinear and/or (2) coupled.

The carbon dioxide adsorption system of the Apollo space cabin life support system has been chosen as the most suitable element of the recovery system for the application of cycling, because the adsorber features a strong thermal field coupled with mass transfer.

2. Progress to date. The major effort of the investigation has been devoted to establishing a satisfactory mathematical model, with a sound basis in theory, for the CO_2 adsorption system behavior. Such a model has been established for mass transfer during adsorption. The following system of equations may be written:

Gas phase: $$\epsilon SC \frac{\partial x_A}{\partial t} = - W \frac{\partial x_A}{\partial z} - Sk \cdot (x_A - x_{A_0}) \qquad (1)$$

Solid phase: $$(1-\epsilon) \cdot S \frac{\partial C_{As}}{\partial t} = Sk \cdot (x_A - x_{A_0}) \qquad (2)$$

The equilibrium relation between solid-phase concentration and interfacial gas-phase concentration of CO_2 is assumed linear:

$$x_{A_0} = mC_{As} \qquad (3)$$

and the respective solutions are given by

$$\frac{x}{x_{A_1}} = 1 - \int_0^{\zeta} \exp(-\tau - \zeta) \cdot I_0(\sqrt{4\tau\zeta})\, d\zeta \qquad (4)$$

$$\frac{mC_{As}}{x_{A_1}} = \int_0^{\tau} \exp(-\tau - \zeta) \cdot I_0(\sqrt{4\tau\zeta})\, d\tau \qquad (5)$$

where I_0 is the Bessel function of the second kind of order zero, and other terms are as defined in the List of Symbols. The data of Figure 1, taken from measurements with a prototype adsorption unit[7], show that the model closely matches the results if a mass transfer coefficient of k = 7 is assumed. This is to be compared with a reported value of k = 5.7 obtained from diagrams reported elsewhere in the same document (in which the present model was not employed). This order-of-magnitude agreement is considered satisfactory in view of several uncertainties in the experimental data of Reference 7.

Computations of CO_2 gas concentration have been performed with the Burroughs B5500 digital computer of the University of Denver, using the infinite-series approximation of equation (4) suggested by Luke[8].

A significant conclusion emerging from this satisfactory model is that axial diffusion makes no significant contribution to equation (1). The heat transfer equations analogous to (1) and (2) have exactly the same form, with heat transfer coefficient h in place of mass transfer coefficient k, temperature as the independent variable in place of concentration, and corresponding thermal capacity factors as coefficients. On the basis of the finding as regards axial mass diffusion, axial thermal diffusion will be neglected in future computations.

During the early stages of this investigation, the principal investigator visited NASA Langley for consultation with Mr. A. O. Pearson's group who are engaged in research on the CO_2 recovery system. This visit resulted in the acquisition of considerable data of direct use to the present investigation, but revealed that thermal data comparable to the concentration data of Figure 1 were not available.

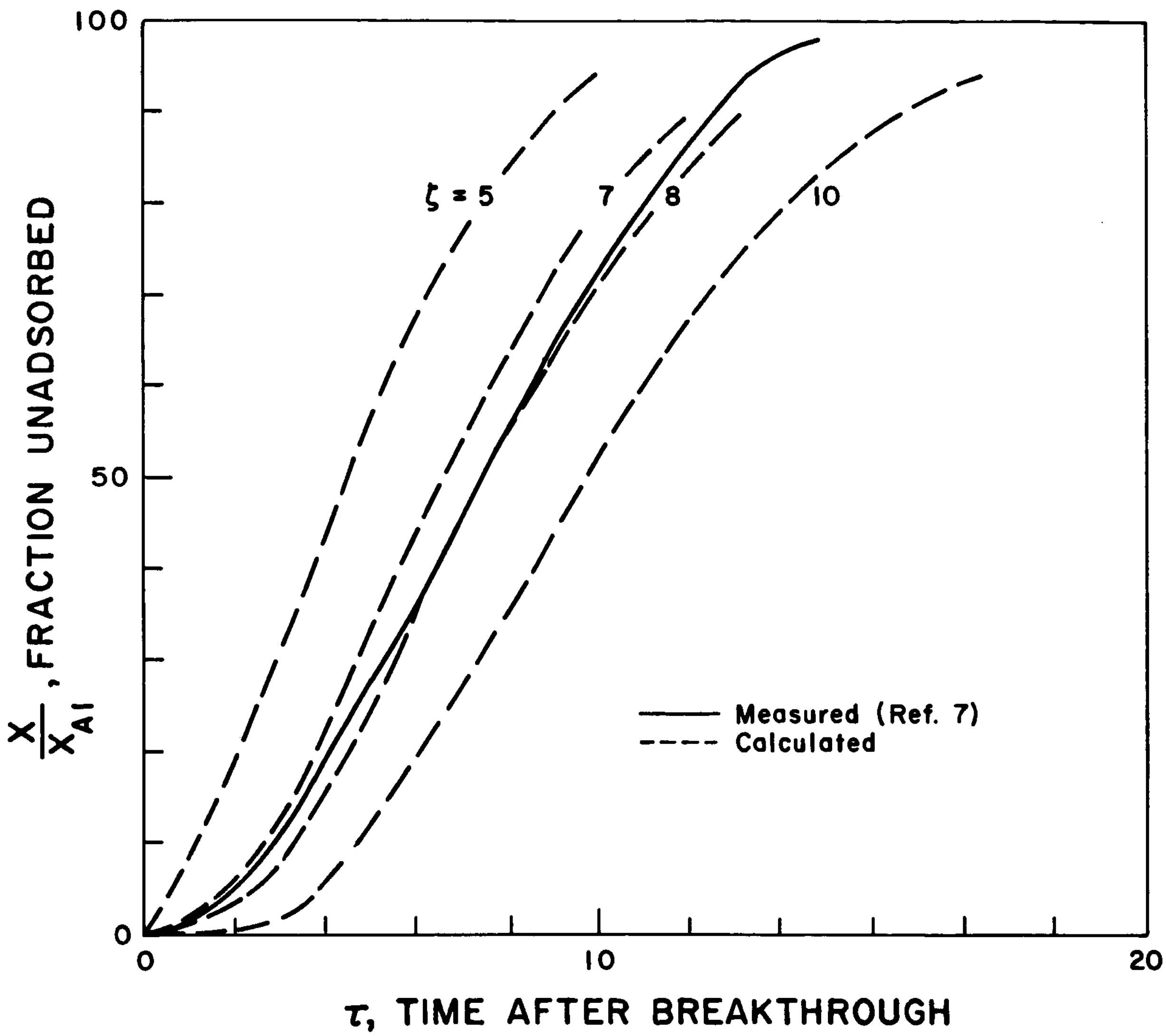


Figure 1. Breakthrough Curves for CO_2 Adsorption Bed

3. <u>Future conduct of the investigation</u>. Construction of an adsorption bed that will permit precise measurements of temperature and CO_2 concentration of the bed effluent is in progress. This experimental bed has approximately the same dimensions as the actual unit, but differs slightly in cooling coil configuration. It is not believed necessary to employ a rigorously identical unit, since an adequate mathematical model is available.

Experiments will be performed in late summer. The results desired from these experiments include

a) concentration-time data similar to Figure 1,

b) temperature-time data at the bed outlet,

c) temperature-time profiles within the bed at various points.

A very accurate thermal conductivity cell is available in the Department of Chemical Engineering for CO_2 concentration measurements, and conventional thermocouples and a millivolt potentiometer will permit highly accurate temperature measurements.

With temperature parameters established, it will then be possible to solve the concentration and temperature equations simultaneously. These equations are linear and coupled. Now the inlet velocity is made periodic (e.g., sinusoidal) and the equations become

$$\epsilon SC \frac{\partial x_A}{\partial t} = - W(t) \cdot \frac{\partial x_A}{\partial z} - Sk \cdot (x_A - x_{A_0}) \tag{6}$$

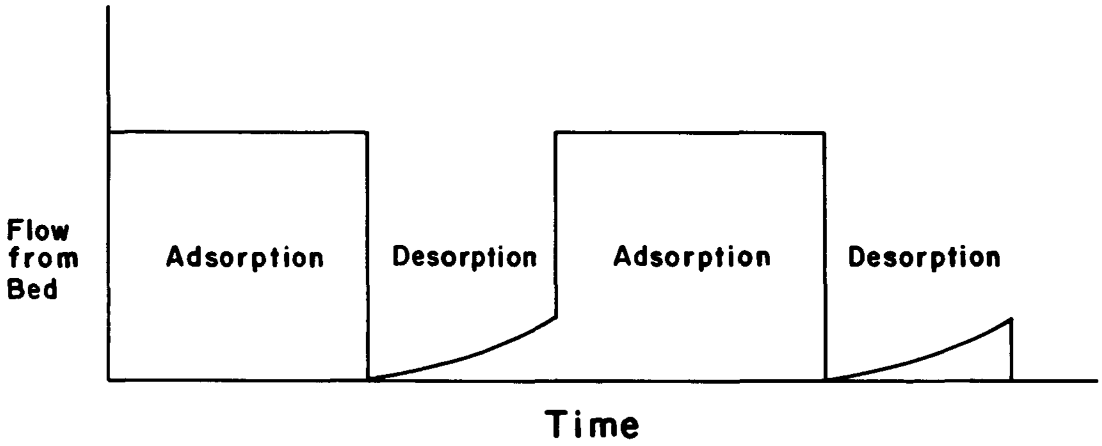
$$(1 - \epsilon) \cdot S \frac{\partial C_{As}}{\partial t} = Sk \cdot (x_A - x_{A_0}) \tag{7}$$

$$\epsilon SC_p \frac{\partial T}{\partial t} = - W(t) \cdot \frac{\partial T}{\partial z} - Sh \cdot (T_s - T) \tag{8}$$

$$(1 - \epsilon) \cdot S \frac{\partial T_s}{\partial t} = Sh \cdot (T_s - T). \tag{9}$$

This coupled system is to be solved for x_A as a function of time, with frequency ω and length of cycle as parameters.

It will be observed that the adsorption bed is normally operated in a square-wave cyclic mode:



The problem of this investigation is to vary the mode of cycling in order to maximize the adsorption bed performance. For the period before CO_2 breakthrough, performance may be expressed by

$$M = \frac{W \cdot x_{A_0} \int_0^{\tau} [1 - \exp(-\zeta)]\, d\zeta}{V_s \zeta} \tag{10}$$

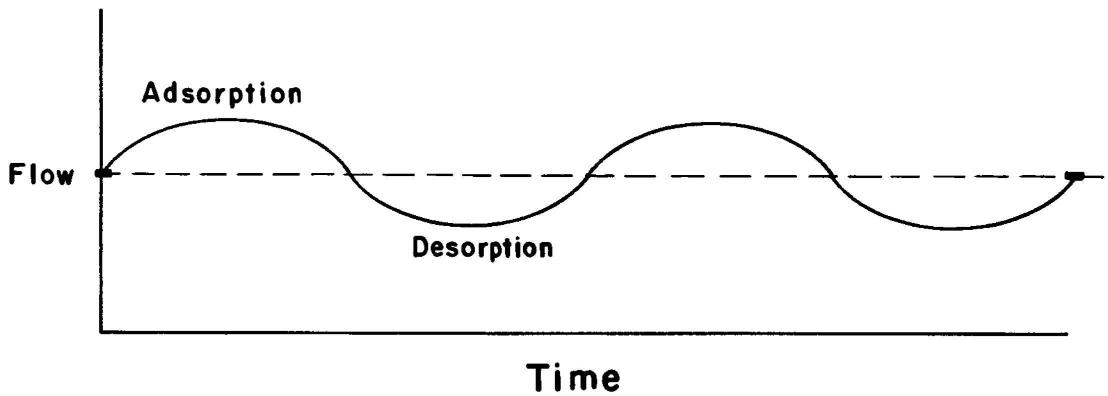
and for the period after CO_2 breakthrough, i.e., for $\tau > 0$,

$$M = \frac{W \cdot x_{A_0} \int_0^{\tau}\int_0^{\zeta} \exp(-\tau-\zeta) \cdot I_0(\sqrt{4\tau\zeta})\, d\zeta d\tau}{V_s \zeta \tau} . \tag{11}$$

Because the integral expressed in equation (11) cannot be obtained in closed form, digital computer methods of integration will be required to determine the behavior of the function M. As a preliminary observation, it seems evident that adsorption-bed performance is maximum at the time of breakthrough.

However, overall performance (adsorption plus desorption) is not necessarily optimum if adsorption is terminated at breakthrough. Desorption data comparable to Figure 1 are required for examination of the complete cycle.

The final phase of the investigation will consist of applying a sinusoidally varying flow rate to the bed as illustrated in the following sketch:



This is not necessarily practical with the present bed design, but the sinusoidal mode may result in the type of coupling of fields that provides enhancement of operation. With two beds in parallel, sinusoidal operation could be conducted such that the respective flows in the two beds is 180° out of phase, continuously varying.

With the present design, sinusoidal cycling is disadvantageous because the CO_2 cannot be concentrated for feed to the reactor. This would become a design problem if the method appeared advantageous from the standpoint of performance.

Sinusoidal cycling will be studied mathematically and experimentally. There is a problem in choosing the degree of heat removal from the adsorption bed, since heat production will vary continuously. This will be resolved on the basis of laboratory findings, expediency, and actual spaceflight requirements.

IV. Project Personnel

Project personnel have included Dr. L. W. Ross as project director, and Mr. F. I. Honea and Mr. L. von Szirmay as graduate research assistants.

Mr. Honea holds an M. S. degree in mechanical engineering from the University of Southern California, specializing in fluid mechanics and heat transfer. He has worked with North American Aviation and Martin-Marietta on a number of aerospace-related assignments. He is presently a Ph. D. candidate in chemical engineering at the University of Denver.

Mr. von Szirmay holds an M. S. degree in chemical engineering from the University of Detroit. He will join the University of Denver chemical engineering faculty as instructor in the fall quarter 1968, and continue work toward the Ph. D. degree. He is a native of Hungary (now a Canadian citizen) and is widely experienced in the chemical industries. At Denver, Mr von Szirmay has been responsible for operation and maintenance of the chemical engineering laboratories, an assignment that he has performed with outstanding success. His Ph. D. thesis topic is a study of hydrocarbon adsorption, closely related in technique to the present investigation.

List of Symbols

C	Total molar concentration of gas phase
C_{As}	Concentration of CO_2 in solid phase
C_p	Molar heat capacity of gas phase
h	Overall heat transfer coefficient
k	Overall mass transfer coefficient
m	Gas-solid equilibrium coefficient, equation (3)
M	Performance function of adsorption bed
S	Cross section of adsorption bed
t	Time
t'	Breakthrough time at arbitrary distance z, defined as $t - z\epsilon SC/W$
T	Temperature of gas phase
T_s	Temperature of solid phase
V_s	Volume of solid phase
W	Molar flow rate of carrier gas
x_A	Mole fraction CO_2 in gas phase
x_{A_0}	Equilibrium mole fraction of CO_2 in contact with solid phase
x_{A_1}	Initial mole fraction of CO_2 in gas
z	Longitudinal distance in adsorption bed
ϵ	Fraction of bed space occupied by gas phase
τ	Dimensionless breakthrough time at arbitrary distance z, defined as $mt'k/(1-\epsilon)$
ζ	Dimensionless distance, defined as zSk/W

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