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Separation of isomers via thermal parametric pumping

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SEPARATION OF ISOMERS VIA THERMAL PARAMETRIC PUMPING

BY

VINCENT J. D'EMIDIO

A THESIS

PRESENTED IN PARTIAL FULFILLMENT OF

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OF

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AT

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ABSTRACT

Continuous and semicontinuous thermal parametric pumps for separating isomers were experimentally investigated using the model system glucose-fructose-water on a cation exchange resin adsorbent. A comparison was made between the experimental data and the calculated results by a method based on an equilibrium theory. The method invokes the assumptions that a multicomponent mixture contains a series of pseudo binary systems. Each binary system consists of one of the solutes as one component and the common inert solvent as the other component. It has been shown that under certain conditions, the pump with feed at the enriched end has the capacity for complete removal of fructose from one product stream and at the same time give arbitrarily large enrichment of fructose in the other product stream.

APPROVAL OF THESIS

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FOR

DEPARTMENT OF CHEMICAL ENGINEERING

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INTRODUCTION

Parametric pumping is a cyclical separation process consisting of a solid phase adsorbent and a fluid phase containing two or more components. In the direct mode of parametric pumping, solution flows through a jacketed glass column packed with adsorbent particles in such a way that its direction alternates periodically. Simultaneously, the temperature of the column changes in phase with the cyclic flow by supplying either hot or cold water to the column jacket. Thus when the column becomes cold, solute adsorbs on the solid, leaving the liquid depleted and the column effluent concentration lowered. When the column is heated on the next half-cycle, solute desorbs, re-enters the solution, and the effluent concentration rises. Because the flow is reciprocating, these effluents occur at opposite ends of the bed. Hence a separation is produced. (Gregory et. al. (13)) The experimenter acts externally on the system to couple these periodic actions by bringing them into synchronism and by establishing a desirable phase angle between them. (Wilhelm et. al. (5); See Appendix, Fig. 1, p.55)

In the recuperative mode of parametric pumping, fixed thermal boundary conditions at the column ends cause the fluid to carry not only the mixture components but also heat into and out of the column as the flow direction alternates. The heat flux alternations establish the parametric temperature variations throughout the column. The combination of flow alternation, the direct thermal contact between phases, and the establishment of heat source-sink positions at the column ends forms an internal recuperative heat exchange system as an integral part of parametric pumping. Such a system has a thermal advantage over the direct mode in which heat is introduced laterally to the flow direction. It is recuperative in that much of the heat which enters the bed during downflow will be in the hot product stream which leaves the bed during upflow, thus allowing for heat recovery and re-use. (Wilhelm et. al. (5)) As in the case of the direct mode, there is coupled interaction of fluid displacement alternations with periodic changes in temperatures and compositions. These events cause the development of an axial flux of adsorbable material and, in turn, formation of an axial separation (Wilhelm et. al. (5); See Appendix, Fig. 2, p.56).

To summarize then, the degree of freedom offered by the independent, direct controllability of the direct mode permits placement of the system in operational domains in which large separations may occur, but at the expense of primary energy (heat) recovery. For many purposes, such recovery may not be important compared to the achievement of separation. By contrast, in the recuperative mode, the periodic actions in question no longer are independent and smaller separations generally may be expected. These are counterbalanced by a recuperative, internal recovery of heat. (Wilhelm et. al. (5))

A distinction is made between closed and open parametric pumping systems in a traditional way. A closed system has no feed or product streams, it operates cycle after cycle with the same volumetric displacement in each direction. (If it were otherwise, all the solution would accumulate at one end of the bed). Open systems require feed input and product removal during part or all of each cycle. In these systems it is not always necessary to restrict the axial fluid displacements to equal volumes on each half cycle. (Gregory et. al. (13))

Both systems becomes steady-periodic as the number of cycles increases, and no further change occurs in the concentrations at the column ends, i.e., a limiting separation is approached. After limiting conditions are reached an open system continues to yield additional product each cycle. A closed system, however, exhibits no change in product volume (Gregory et. al. (13)).

TABLE 1

CHRONOLOGICAL SUMMARY OF THE CYCLIC SEPARATION METHODS REVIEWED IN THE INTRODUCTION

YEAR	AUTHOR(S)	INTENSIVE THERMODYNAMIC VARIABLE	SYSTEM	REMARKS
1966	Wilhelm, et. al. (1)	Temperature	$NaCl$-H_2O on Rohm and Haas Amberlite Resins (IRC-50, IR-45)	Experimental and Mathematical Model
1967	McAndrews (2)	Temperature	Methane-Nitrogen on Fibrous Carbon	Experimental and Mathematical Model
1968	Jenczewski, et. al. (3)	Temperature	Ethane-Propane on Activated Carbon	Experimental
1968	Wilhelm, et. al. (4)	Temperature	Toluene-n-Heptane on Silica Gel	Experimental and Mathematical Model
1968	Wilhelm, et. al. (5)	Temperature	A. Toluene-n-Heptane on Silica Gel B. $NaCl$-H_2O on Rohm & Haas Amberlite IR-45 and IRC-50 Resins	Experimental and Mathematical Model
1969	Rolke, et. al. (6)	Temperature	$NaCl$-H_2O	Mathematical Model
1969	Pigford, et. al. (7)	(Thermal)	-	Mathematical Model
1969	Aris, (8)	(Thermal)	-	Mathematical Model
1969	Sweed, et. al. (9)	(Thermal)	-	Mathematical Model
1969	Horn, et. al. (10)	(Thermal)	-	Mathematical Model

TABLE 1

CHRONOLOGICAL SUMMARY OF THE CYCLIC SEPARATION METHODS REVIEWED IN THE INTRODUCTION

YEAR	AUTHOR(S)	INTENSIVE THERMODYNAMIC VARIABLE	SYSTEM	REMARKS
1969	Pigford, et. al. (11)	Temperature	A. Methane in Helium on Pittsburgh BPL Activated Carbon B. Acetic Acid-H_2O on Pittsburgh CAL Activated Carbon	Experimental
1970	Sabadell, et. al.	pH	K^+ and Na^+ - H_2O on Rohm & Haas IRC-84 a carboxylic polyacrylic resin	Experimental
1970	Gregory, et. al. (13)	(Thermal	-	Mathematical Model
1970	Jenczewski, et. al. (14)	Temperature	A. Argon-Propane on Pittsburgh BPL Activated Carbon B. Ethane-Propane on Pittsburgh BPL Activated Carbon C. Propane-Propylene on Pittsburgh BPL Activated Carbon	Experimental and Mathematical Model

TABLE 1

CHRONOLOGICAL SUMMARY OF THE CYCLIC SEPARATION METHODS REVIEWED IN THE INTRODUCTION

YEAR	AUTHOR(S)	INTENSIVE THERMODYNAMIC VARIABLE	SYSTEM	REMARKS
1971	Chen, et. al. (15)	(Thermal)	-	Mathematical Model
1971	Sweed, et. al. (16)	Temperature	N_aC_l-H_2O on Bio-Rad AG 11A8 Resin	Experimental and Mathematical Model
1971	Turnock, et. al. (17)	Pressure	Nitrogen-Methane on Linde Type 5A Molecular Sieve	Experimental and Mathematical Model
1971	Gupta, et. al. (18)	(Thermal)	-	Mathematical Model
1971	Baker, et. al. (19)	Temperature	Acetic Acid-H_2O on Activated Carbon	Experimental
1972	Butts, et. al. (20)	(Thermal)	-	Mathematical Model
1972	Gregory, et. al. (21)	Temperature	N_aC_l-H_2O on Bio-Rad AG 11A8 Resin	Experimental and Mathematical Model
1972	Chen, et. al. (22)	Temperature	Toluene-n-Heptane on Silica Gel	Experimental and Mathematical Model
1972	Chen, et. al. (23)	Temperature	Glucose-Fructose-H_2O on Fullers Earth	Experimental
1972	Kowler, et. al. (24)	Pressure	Nitrogen-Methane on Davison 5A Molecular Sieve	Experimental and Mathematical Model

TABLE 1

CHRONOLOGICAL SUMMARY OF THE CYCLIC SEPARATION METHODS REVIEWED IN THE INTRODUCTION

YEAR	AUTHOR(S)	INTENSIVE THERMODYNAMIC VARIABLE	SYSTEM	REMARKS
1972	Shendalman, et. al. (25)	Pressure	Carbon Dioxide in Helium on Silica Gel	Experimental and Mathematical Model
1972	Patrick, et. al. (26)	Temperature	Air-SO_2 on Silica Gel	Experimental
1973	Chen, et. al. (27)	Temperature	Toluene-n-Heptane on Silica Gel	Experimental and Mathematical Model
1973	Butts, et. al. (28)	Temperature	K^+ - H, and K^+ - Na^+ H^+ on Dowex 50 x 18 Resin	Experimental and Mathematical Model
1973	Wankat, et. al. (29)	Temperature	Acetic Acid-Water-Diethyl Ether	Experimental and Mathematical Model
1973	Gupta, et. al. (30)	(Thermal)	-	Mathematical Model
1973	Mitchell, et. al. (31)	Pressure	CO_2-Helium on Silica Gel	Experimental and Mathematical Model
1974	Apostolopoulos (32)	Temperature	-	Mathematical Model
1974	Chen, et. al. (33)	Temperature	Toluene-n-Heptane on Silica Gel	Experimental
1974	Chen, et. al. (34)	Temperature	Toluene-Aniline-n-Heptane on Silica Gel	Experimental and Mathematical Model

TABLE 1

CHRONOLOGICAL SUMMARY OF THE CYCLIC SEPARATION METHODS REVIEWED IN THE INTRODUCTION

YEAR	AUTHOR(S)	INTENSIVE THERMODYNAMIC VARIABLE	SYSTEM	REMARKS
1974	Chen, et. al. (35)	Temperature	N_aNO_3-H_2O on Bio-Rad Ag 11A8 Resin	Experimental
1975	Chen, et. al. (36)	Temperature	Glucose-Fructose-H_2O on Bio-Rad AG 50W-X4 Resin	Experimental

TABLE 2*

REQUIREMENTS FOR PARAMETRIC PUMPING

	Conventional Phase Exchange Processes	Parametric Pumping (General)
Number of Phases	At least two	At least two
Equilibrium Function	Any function	Any function
Relative Motion Between Phases	Unidirectional	Alternating
State Variables	All steady	One or more forced periodically. Others periodic in response.

*Sweed, et. al. (9)

Systems Based on Temperature Cycles

Thermal parametric pumping was conceived by Wilhelm about 1962. In the paper of Wilhelm, et. al. (1) parametric pumping is initially described and its principles initially presented via the recuperative mode of operation for both open and closed systems. For the open systems where fresh feed was introduced each half-cycle, the removal of NaCl from water on a mixed bed of ion exchange resins was studied. A separation factor (defined as the ratio of concentration of solute in the upper reservoir to concentration of solute in the lower reservoir) of 1.2 was obtained. As Wilhelm, et. al. (1) describes it, alternating axial displacement of a fluid mixture in a column of adsorptive particles upon which an axial temperature difference is imposed leads, through coupling of oscillatory thermal and mass fields with alternating flow displacements, to a difference in limiting condition, time-average compositions at the column ends. The separation takes place at the expense of thermal energy; continuous operation with a fixed-bed adsorber also becomes feasible. The separation has been substantiated by experiment and by analysis of the mathematical formulation.

McAndrew (2) provided the first reported work on the use of parametric pumping principles to separate gases. McAndrew (2) used the recuperative mode of operation to separate methane from nitrogen an fibrous carbon via batch parametric pumping. The mathematical model revealed some problems. Under normal operations a big difference between solid and gas heat capacities makes it difficult to use the recuperative mode for heating the bed, also the fundamental P-V-T behavior of gases demands that when the temperature changes a corresponding change in either volume or pressure must occur. So at constant pressure operation, the volume increases, dilutes the gases and separation drops. If the operation is at constant volume, the increasing pressure forces increasing adsorption. However this action is opposite to the tendency of a drop in adsorption which an increasing temperature effects. McAndrew (2) did not obtain any experimental separations.

Jenczewski, et. al. (3) were the first to employ the direct mode to the separation of gases. The separation of ethane-propane on activated carbon was studied in a constant volume apparatus for a closed system. Separation factors of approximately 2.0 were obtained following 60 cycles of pump operation. A horizontal column was utilized in which the flow periods were fast, with longer periods in order to attain equilibrium.

Wilhelm, et. al. (4) employed the direct mode to the separation of toluene-n-heptane on silica gel. The differential mass balance and an approximate rate expression were presented. Theory is in qualitative agreement with the experimental results. As Wilhelm, et. al. (4) puts it, parametric pumping, a dynamic separation technique, comprises alternating axial displacement of a fluid mixture in a column of adsorptive particles upon which a synchronous cycling temperature is imposed. With direct coupling, separations of 10^5:1 between column ends have been obtained with indications of great potential for further separation. A mathematical model and its solution describe the behavior of the system.

Wilhelm, et. al. (5) employed the direct mode to the separation of toluene-n-heptane on silica gel and the recuperative mode to the separation of $NaCl-H_2O$ in Rohm and Haas amberlite IR-45 and IRC-50 resins. The theory presented before (4) is also presented in greater detail. Wilhelm, et. al. (5) were reasonably successful in predicting the separation achieved. According to Wilhelm, et. al. (5), parametric pumping is an adsorptive separation technique based on periodic, synchronous, coupled transport actions. The net consequence of the coupling of alternating adsorbent-fluid displacements with the cycling of a thermodynamic intensive parametric variable (temperature) is a buildup of separation from cycle to cycle. Thereby the powerful separational effects of countercurrent action are released in the uncommon circumstances of a continuously regenerating separation column having only a single fluid phase. Thus cycle-to-cycle time-staging as well as position-staging occurs within a column. Parametric pumping also is a compound, macroscopic, active transport system in which a species is moved at the expense of some form of energy (thermal, chemical potential) from a region of low concentration to high. The large separation capability of the direct mode of parametric pumping is demonstrated experimentally for a toluene-n-heptane mixture on silica gel. A very important theoretical phase-angle relationship between oscillatory parts of the system has been verified experimentally for this mode. New results are presented for the recuperative mode in which heat is exchanged internally. $NaCl-H_2O$ was separated in a continuous open system on ion exchange resins. Preliminary theoretical efficiencies have been computed. The separation factors were predicted to increase gradually with each thermal-flow cycle and to be limited finally only by the mass transfer resistance to diffusion of material between solid and fluid phases and by axial diffusion.

Rolke, et. al. (6) used the experimental data of (5) for the $NaCl-H_2O$ system and developed precise modeling of open recuperative mode parametric pumping without any recycle (1,5). As Rolke, et. al. (6) explains, a computational model, based on finite difference solution of partial differential equations, is developed to describe thermal and solute concentration behavior within a recuperative parametric pumping column. Experimental data for the dilute NaCl solution-mixed bed ion exchange resin system are used to evaluate parameters in the model, which successfully simulates the time-and position-varying column profiles during an experimental NaCl-Water separation run. To model accurately this system in which interphase solute transfer is strongly intraparticle diffusion controlled and alternating in direction, intraparticle profiles must be taken into account. A mean separation factor of 1.11 and a maximum separation factor of 1.22 were obtained.

Pigford, et. al. (7) present the most easily interpreted mathematical model of the direct mode thermal parametric pump, called the equilibrium theory. Their development of this model pertains to special conditions for the batch operation of a parametric pump. According to Pigford, et. al. (7), very large separation factors have been obtained by Wilhelm and his co-workers using cycling flow of a binary mixture upward and downward through a column containing a fixed bed of solid adsorbent which is alternately heated and cooled. The theory of such separations is developed here on the assumption of local equilibrium between solid and fluid phases. The origin of the separation is the ability of the solid phase to store solute deposited on it by fluid flowing from the bottom of the column and to release this solute later into another fluid stream which flows into the column from a top reservior containing enriched mixture. The proposed mechanisim takes into account the difference in the speeds of propagation of concentration waves through the packing during upward and downward flow. The assumptions made here simplifies the equations because no rate equations need be employed. Also, the equilibrium relationship was assumed linear, and axial dispersion was neglected. Pigford, et. al. (7) also fit their theory to the data obtained by Wilhelm, et. al. (4), which was accomplished by fitting only one parameter, the equilibrium constant change parameter b, to the data. Doing this enables b to include other effects such as dispersion and rate limitations which occur in the column and won't agree with the value obtained from equilibrium data. The expressions for the top and bottom composition are:

$$(Y_t)n = Yo \left(2 + \frac{2b}{1+b} - \frac{(1-b)}{(1+b)}^{n-2} \right) \quad ; n > 2 \qquad \text{(A)}$$

$$(Y_t)n = Yo \quad \frac{(1-b)}{(1+b)} \qquad \text{(B)}$$

If we define a separation factor $S=(Y_t)n \;/\; (Y_t)n$ then

$$S_n = \left((2 + \frac{2b}{1+b}) \right) \left(\frac{(1+b)}{(1-b)} \right)^n - \frac{(1+b)}{(1-b)}^2 \quad ; n > 2 \qquad \text{(C)}$$

(Patrick, et. al. (26))

Aris (8) showed that the equilibrium theory of Pigford et. al. (7) is a special case of a more general theory and derived the general theory.

Sweed, et. al. (9) presented a computational investigation of separation by direct mode liquid phase parametric pumping. This is the STOP-GO algorithm. They have simulated experimental separations presented previously by the authors (4). According to Sweed, et. al. (9), a computational investigation of separation by direct thermal mode, liquid phase parametric pumping is presented. Calculations are performed using the new STOP-GO algorithm, a modification of the method of characteristics. Using the toluene-n-heptane-silica gel system, they have determined the effect of displacement, cycle time, phase angle, and reservoir volume on separations. Separation factor increases exponentially with number of cycles, and in the case of equilibrium operation, there is almost no limit to separation capability. A graphical calculation procedure also is presented for equilibrium operation. The STOP-GO model is more realistic than the equilibrium model, however numerical computer calculations are required.

Horn, et. al. (10) studies were solely concerned with open systems. They provided the first detailed presentation with theoretical calculations via direct mode parametric pumping. Horn, et. al. (10) studied a parametric pump setup with center feed into a central reservoir and product takeoff from the two end reservoirs. This paper, however, is presented in very general terms without any detailed examples or experimental data.

Pigford, et. al. (11) employed the direct mode to the separation of a gas-solid system, methane in heluim on Pittsburgh BPL activated carbon and a liquid-solid system, acetic acid-water on Pittsburgh CAL activated carbon, via a modified version of parametric pumping called cycling zone adsorption. As Pigford, et. al. (11) relates, cyclic changes in concentration are produced in a fluid which flows through a fixed bed of solid adsorbent owing to temperature cycling of the bed. The product stream is collected separately during the periods of positive and negative deviations from the feed composition. The separation is increased by using separate zones in series, the temperature changes in adjacent zones being out of phase. The separation is governed by the wave propagation properties of the beds. This process resembles parametric pumping except that the fluid to be separated flows in a single direction through an arbitrary number of columns, instead of back and forth through a single bed. What is termed the standing wave mode of operation is the same as the direct mode of parametric pumping and what is termed the traveling wave mode of operation is the same as the recuperative mode of parametric pumping. Initial results from four experiments, a single-zone traveling wave separation of acetic acid from water, a single-zone standing wave separation of methane from helium, and single and double zone standing wave separations of acetic acid from water were presented. The two-zone system yielded greater separation capability, and the traveling wave separation proved to be better than the standing wave separation for a single zone.

Gregory, et. al. (13) extended the equilibrium theory of Pigford, et. al. (7) to include reservoir dead (void) volume. Their studies were concerned with both open and batch systems via direct mode parametric pumping. According to Gregory, et. al. (13), an analytical solution of the model equations which describe continuous direct thermal parametric pumping systems is presented. Although the solution is strictly applicable only in the limit of equilibrium operation, it does provide insight into the qualitative behavior of nonequilibrium systems. It is shown how this analytical solution can be used quantitatively for comparing various open system configurations and operations. Earlier work on batch systems is extended to include the effect of reservoir dead volume. After modeling two open systems, three regions were discovered that related to the bottom product concentration. However, there is only one region which has a limiting concentration of zero. To achieve this limiting concentration of zero condition a constraint must be satisfied, i.e., the bottom reflux ratio (which by definition is the ratio if amount of bottom product per cycle to bottom reservoir displacement) must be greater than a limiting value which depends on the equilibrium constants.

Jenczewski, et. al. (14) employed the direct mode of parametric pumping to the separation of the gases argon-propane on Pittsburgh BPL activated carbon and, propane-propylene on Pittsburgh BPL activated carbon via a batch, constant volume system. The first two gas systems were successfully separated, but the third gas system was not. As Jenczewski, et. al. (14) relate, a closed, thermal, pulsed adsorber was studied. Separations of gaseous mixtures were obtained experimentally for argon-propane and ethane-propane mixtures. No separation was observed for a propane-propylene mixture. An equilibrium model, the parameters of which are measured independently, provides good agreement with the experimental results. This was achieved by altering the STOP-GO model (9) to include axial mixing of the gases effected by temperature fluctuations in the adsorbent bed.

By extending the equilibrium theory of Pigford, et. al. (7), Chen, et. al. (15) have derived mathematical expressions for the performance of batch, continuous, and semicontinuous parametric pumps. Their continuous pump, characterized by a steady flow for both feed and product streams during the upflow and downflow cycles, has a truly continuous operation in nature. On the other hand, the semicontinuous pump is operated batch-wise during upflow and continuously during downflow. They have shown that under certain conditions the batch pump and the continuous and semicontinuous pumps with feed at the enriched end have the capacity for complete removal of solute from one product stream and, at the same time, give arbitrarily large enrichment of solute in the other product stream. Furthermore, they have found with experimental verification that depending upon the relative intensities of certain parameters, three distinct regions of separation exist for the three modes of pump operation. One region is characterized by the eventual and complete removal of solute at one product stream with a certain level of solute enrichment present at the other product stream. The other two regions are characterized by only partial removal of solute from one product stream with some solute enrichment at the other end. Since in most cases complete or nearly complete removal of solute is desired, the operating parameters of the pump should be adjusted in such a way so as to insure operation in the proper region of separation.

Sweed, et. al. (16) used the STOP-GO method to simulate new data on the separation of NaCl-Water on Bio-Rad AG 11A8 ion retardation resin via batch, direct mode (including reflux) parametric pumping. According to Sweed, et. al. (16), direct thermal parametric pumping separation of a NaCl-Water ion retardation resin system have been investigated. Experimental breakthrough curves and batch parametric pumping runs were used to determine the lumped-parameter mass transfer coefficient and its dependance on velocity and temperature. Computer simulation of additional batch runs verified both the model and the STOP-GO algorithm used to solve it. Small separation factors close to 10 were obtained.

Gupta, et. al. (18) independently developed an equilibrium theory using the method of characteristics for direct mode (standing wave) cycling zone adsorption. According to Gupta, et. al. (18) cycling zone adsorption is a separation process in which fluid is passed through a series of adsorbent zones, the periodic temperature changes in adjacent zones being one-half cycle out of phase with one another. This paper presents a method for computing the separation as a function of fluid displacement, cycle time, and adsorptive equilibrium parameters. Criteria are developed for optimum separation. This analysis is restricted to linear isotherms and instantaneous local equilibrium. The results of the computer model revealed that finite mass transfer rates and axial dispersion prevent the occurence of an infinite separation.

Baker, et. al. (19) presented both experimental and theoretical aspects of the separation of Acetic Acid-Water on activated carbon via cycling zone adsorption. As Baker, et. al. (19) relates, a theoretical explanation for cycling zone adsorption, a wave propagational separation process given a qualitative explanation in an earlier article (11), is presented. Experimental results confirming the theoretical predictions are also included. It is found that in addition to accounting for the separation effect the theory predicts the effect can be amplified through the interaction of the concentration and progressing thermal waves. Possible process schemes utilizing the interactions are discussed. One and two zone standing and traveling wave modes were applied to the model system. Somewhat better separations were obtained for the traveling wave mode. Agreement between theory and experiment was fairly good considering that the necessary equilibrium parameters were obtained from equilibrium experiments and not from a mathematical fit of the experimental runs.

Butts, et. al. (20) extended the equilibrium theory of Pigford, et. al. (7) to study separations of multicomponent mixtures via direct mode, batch parametric pumping using unsymmetrical cycles with greater flow to one of the reservoirs. As Butts, et. al. (20) explains, a new parametric pump process is described in which it is possible to cause certain solutes to migrate upward in a chromatographic column while the other solutes migrate downward at the same time, thereby effecting separation. This new process uses a nonsymmetric flow pattern in the direct thermal mode operation. Separation can be predicted from algebraic equations developed under the assumptions of linear isotherms, no axial dispersion, and instantaneous local equilibrium. Open systems would probably effect better separation because an actual batch process would require big reservoir dead (void) volumes to enable sufficient cycling for separation before all of the liquid was pumped to the bottom reservoir.

Gregory, et. al. (21) considered the separation of NaCl-Water on Bio-Rad AG 11A8 ion retardation resin by continuous, direct mode parametric pumping where dispersive effects are significant. They mathematically modeled four open systems and one batch system using the STOP-GO theory. According to Gregory, et. al. (21), experimental separations of sodium chloride-water mixtures are presented for two distinct modes of continuous, direct thermal parametric pumping. These continuous separations are accurately simulated using a computer solution of the PDE (partial differential equation) model parameters which are obtained entirely from batch experiments. Fives modes of operation are optimized and parameter sensitivity is explored. The STOP-GO model fit the data very nicely while the equilibrium theory (13) predicted greater separation than they found experimentally. A separation factor of nearly 2 was obtained when the equilibrium theory predicted infinite separation factors. This paper lucidly shows that equilibrium theory results should be utilized with circumspect, and that the STOP-GO algorithm shall predict experimental open system separations when the model parameters are obtained from batch parametric pumping experiments.

Chen, et. al. (22) employed the direct mode to the separation of toluene-n-heptane on silica gel via continuous parametric pumping with top feed. In this paper a continuous pump in which the feed and product streams flow steadily both in upflow and downflow is experimentally investigated in the model system toluene-n-heptane on silica gel adsorbent. The experimental data compare reasonably well with analytical results based on an equilibrium theory. The operating conditions necessary to achieve high separation factors are shown to be dependent on the relative magnitudes of penetration distances and the height of the column. In the region where the equilibrium theory predicts infinite separation factors, a separation factor of better than 600 was obtained for 14 cycles of experimental pump operation. Furthermore, more separation could be obtained with more cycles. When the pump operated in the other regions separations curtailed. By fitting the data of the run with the greatest separation, the equilibrium parameters were obtained. Again some of the nonequilibrium column effects will be incorporated in the values of the equilibrium parameters when this procedure is employed.

Chen, et. al. (23) employed the direct mode of parametric pumping to the multicomponent separation of fructose-glucose-water on fullers earth via continuous parametric pumping with top feed. The data, however, only agrees qualitatively with what the equilibrium theory predicts. Application of the Langmuir equation yielded pure component equilibrium isotherms. The two sugars demonstrated notable differences in both their respective isotherms and their repsective equilibrium change parameters, b. Because it took two hours for the system to reach equilibrium,six hour cycle times were required when running. Due to the technique of pump operation, glucose should be completely removed from the bottom product stream and fructose only partially. From the reported b values, the separation may be approximated from the equilibrium theory (22) as being significantly larger than the experimental results revealed.

Patrick, et. al. (26) employed the direct mode to the separation of Air-SO_2 on silica gel via constant pressure, batch operation. Since the operation was at constant pressure, axial flow to or from one of the reservoirs must occur whenever the temperature changed. A limiting separation factor of nearly 130 was obtained, while the equilibrium theory (7) predicted larger separation. As the authors point out, the equilibrium theory does not consider the axial flow caused by the pressure changes. A model involving the axial flow should have been employed instead. The authors also indicate that pressure difference probably has more promise than temperature difference as the thermodynamic intensive variable in parametric pumping separations of gases.

Chen, et. al. (27) employed the direct mode of parametric pumping to the separation of toluene-n-heptane on silica gel via semicontinuous parametric pumping with top feed. In this paper a semicontinuous parametric pump with batch operation during one-half cycle and continuous operation in the other half-cycle was experimentally investigated using the model system toluene-n-heptane on silica gel adsorbent. A mathematical model based on an equilibrium theory is presented and is found to be in good agreement with the experimental results. Furthermore, it is shown that when the penetration distance for the cold cycle is less than or equal to that of the hot cycle and the height of the column, the rate of production of pure solvent by this pump may become quite large compared to the rate of production by a pump in which operation is continuous during both half-cycles. The values of the equilibrium parameter discovered before (22) were utilized here. Since there was not that good of an agreement between the bottom product concentrations obtained here and those obtained previously (22), the implication is that the measured equilibrium parameters incorporate some column effects. Also, the semicontinuous mode of pump operation will have greater flow rates at the same bottom product rate.

Butts, et. al. (28) employed the direct mode to the separation of multicomponent mixtures of cations K^+-H^+ and K^+-Na^+-H^+ on Dowex 50 x 8 resin via batch equilibrium parametric pumping. According to Butts, et. al. (28), direct, thermal parametric pumping has been used to fractionate experimentally K^+-H^+ and K^+-Na^+-H^+mixtures using Dowex 50 x 8 as the ion exchanger. The binary exchange equilibrium is influenced by temperature so that desorption of K^+ and adsorption of H^+ occur simutaneously on heating. K^+ accumulates in the top reservoir with separation factors exceeding 2000:1, while H+ accumulates in the bottom with separation factors exceeding 2000:1 in the opposite direction. In a ternary exchange experiment, the K^+ separation factor was 52,000:1, accumulating in the top, H^+ was 97,000:1 in the opposite direction, and Na^+ almost completely disappeared from both reservoirs. The influence of resin swelling due to temperature is investigated with an equilibrium theory model. This paper seems to present a new and unexpected use of parametric pumping for complete fractionation of ionic mixtures via direct thermal mode operation.

Wankat (29) employed the direct mode and utilized two equilibrium stage theories to analyze liquid-liquid extraction parametric pumping via batch operation. As Wankat (29) explains, parametric pumping is extended to liquid-liquid extraction. Equilibrium staged theories are developed for a parametric pump with discrete transfer and equilibrium steps. Experimental results for separations of acetic acid from water using diethyl ether as a solvent are given for both a continuous flow helix and for a discrete transfer test tube system. The theoretical results predict that large separations can be achieved, but the separation factor never becomes infinite. The experimental separations achieved were less than the theoretically predicted separations but they followed the theoretically predicted trends. The qualitative prediction of the staged model follow the STOP-GO model (9) predictions. However, extraction parametric pumping does not seem to hold the promise that adsorption or ion exchange applications have. For one thing, temperature usually has negligible effect on the distribution coefficients, and for another, a cyclic system would have to rival a steady-state countercurrent system. Extraction parametric pumping does have an advantage in that no solvent recovery step is necessary.

Gupta, et. al. (30) employed a mixing cell model to simulate non-equilibrium parametric pumping via direct mode, batch operations. The equilibrium theory originally proposed by Pigford, et. al. (7) assumes the existence of local equilibrium, linear adsorption isotherms, an no axial diffusion. However all real systems do have axial diffusion and finite mass transfer rates, therefore, the equilibrium theory should not be used for design. As Gupta, et. al. (30) remarks, the mixing cell model of a packed bed is used to analyze non-equilibrium effects in linear parametric pumping. This model yields results which are more realistic than the equilibrium theory. Cyclic steady-state concentrations are determined directly without calculating the transient separations. For close to equilibrium operation, the cell model is simplified to obtain a very fast algorithm of computing parametric pumping separations. Examples are discussed for both batch and continuous multicomponent separation systems. Solutions of the cell model equations were obtained utilizing either matrix exponentiation or Laplace transforms. When the experimentally discovered mass transfer coefficients were utilized, the experimental data of Sweed, et. al. (16) fit very well.

Apostolopoulos (32) employed a perturbation approach in his study of near-equilibrium parametric pumping via the direct, thermal parametric pump as a chemical reactor. The reaction under study was a reversible catalytic reaction which needed adsorption before reaction and then desorption. The thought was that the parametric pump would increase reaction conversion and concentrate the product. Comparison of these theoretical results with results obtained from other theories in limiting cases was not done. He presented four examples but they were not applicable to a real system.

Chen, et. al. (33) essentially presented an experimental continuation of previous papers (22, 27), where the direct mode was employed to the separation of toluene-n-heptane on silica gel via continuous and semicontinuous parametric pumping. The significant runs revealed that smaller values of the equilibrium change parameter b resulting from smaller changes in temperature yielded less separation. Also, as long as cycle time length accomodates the time required to attain equilibrium, then it is important.

Chen, et. al. (34) employed the direct mode to the multicomponent separation if toluene-aniline-n-heptane on silica gel via continuous parametric pumping with top feed. In this paper a thermal continuous parametric pump for separating multicomponent mixtures was experimentally investigated using the model system toluene-aniline-n-heptane on silica gel adsorbent. A simple method for predicting separations is presented and is found to be in good agreement with the experimental results. The method, based on an equilibrium theory, invokes the assumption that a multicomponent mixture contains a series of pseudo binary systems. Each binary system consists of one of the solutes as one component and the common inert solvent as the other component. The equilibrium change parameters, b, were 0.15 and 0.31 for toluene and aniline respectively.

Each b value was obtained by fitting the binary separations. Agreement between experiment and theory equaled that found before (22, 27, 33).

Chen, et. al. (35) employed the direct mode to the separation of sodium nitrate-water on Bio-Rad AG 11A8 ion retardation resin via continuous and semicontinuous parametric pumping. In this paper, Chen, et. al. (35) examined the conditions required to obtain the optimal performance of continuous and semicontinuous parametric pumps via direct mode operations, defined by achieving of the maximum possible solute free product for a given column height. The model system sodium nitrate-water on an ion retardation resin adsorbate was employed. Equations defining maximum values of certain important parameters such as the bottom product flow rate and reservoir displacement rate were developed by extending previous theories of parametric pumping. Experimental runs were carried out using parameters whose value were both less than and greater than the maximum values predicted for optimal performance. It was found that for pumps operating with parameter values less than the limiting maximum values, complete removal of solute from the bottom of the column was obtained, and for those pumps for which the parameters values were greater than the predicted maximum quantities only partial removal of solute was observed. The experimental results agreed quantitatively as well as qualtitatively with the theory for almost all runs.

In this paper Chen, et. al. (36) extended direct mode continuous and semicontinuous parametric pumping to the separation of isomers.

The model system studied is glucose-fructose-water on a cation exchange resin adsorbent, calcium form. A comparison is made between the experimental data and the calculated results based on the method proposed by Chen, et. al. (34). The two sugars are optically active isomers, thus when a polarimeter cell is filled with a solution of the two sugars and placed in a polarimeter, glucose will rotate the analyzer to the right (Dextro-rotatory) and fructose will rotate the analyzer to the left (Levo-rotatory). Though glucose and fructose have identical molecular weights their optical rotation is different and characterizes respective component concentrations in a particular solution. It should be pointed out that sugar mixtures are notoriously difficult to separate (Hatt, et. al. (39)) and there has never been a thorough study done on the separation of glucose and fructose, even though it might be advantageous industrially in the manufacture of fructose from sucrose via invert sugar.

Systems Based on Pressure Cycles

Heatless adsorption or pressure swing adsorption was invented by Skarstrom (37). In this process adsorption of solute from a gaseous stream occurs at high pressure while desorption occurs at low pressure. Utilizing some of the high-pressure product stream, the bed may then be purged at low pressures. In his original demonstration of the process, Skarstrom (37) used two columns, one column adsorbing at high pressure while the gas flows up the column, and a second column would be desorbing at low pressure while the gas flows down the column. The beds would interchange after the passage of a few minutes. Skarstrom (37) however did not present any theoretical analysis.

Turnock, et. al. (17) employed pressure changes for the separation of methane from nitrogen on a Linde type 5A Molecular sieve via periodic adsorption operation. As Turnock, et. al. (17) explains, the periodic process utilizes a rapid pressure swing cycle in an adsorbent bed to effect the separation of gas mixtures. During the first portion of a cycle the compressed gas mixture flows into the adsorbent-filled column. Next, while the feed gas is restrained, an exhaust orifice is opened at the feed end of the column providing depressurization. The product stream is enriched in the component exhibiting the lowest coefficient of adsorption. A mathematical model based upon the assumption of instantaneous equilibrium between the gas phase and the adsorbed gas was formulated and solved to simulate the periodic, adsorption process. The measured nitrogen content of the product gas stream was found to correlate with the ratio of the product gas rate to the feed gas rate. At 24^0C the calculated pressure response, feed gas flow rate, the product gas composition correspond favorably with related experimental measurements for all values of the feed gas pressure, cycling frequency, and product gas flow rate within the ranges investigated. Turnock, et. al. (17) also operated at -77^0C and when they did, significantly less separation was obtained when the equilibrium theory predicted greater separation. Apparently rate processes become important when working at -77^0C. Turnock, et. al. (17) also noticed a drop in flow capacity because of molecular sieve attrition. Utilization of hard spheres (24) proved to be the solution to this problem.

Kowler, et. al. (24) again employed pressure changes for the separation of methane from nitrogen via periodic adsorption operation, and discovered that there was an optimum cycle time of nearly three seconds. According to Kowler, et. al. (24) cyclic pressure variations in a fixed bed adsorber can cause significant separation of gaseous mixtures. Feed pressure changes are the driving force in this parametric pumping process. The optimal feed sequence is maximum (maximum flow in) pressure, zero flow (variable pressure), maximum flow out (minimum pressure). For the nitrogen-methane feed gas at $168kN/M^2$ and a 1.22m bed of adsorbent, the optimal cycle time is 3 seconds, and feed is sustained 50% of the time. The zero flow mode is unnecessary if product purity is the sole objective. Also, the fixed bed binary gas adsorber when alternately fed and exhausted at one end, produces a purified product from the other end. Coupled partial differential equations in pressure and composition, representing total mass and component balances with local equilibrium describe the operation. The Maximum Principle is applied to determine the optimal cyclic, unsteady feed policy for the balanced objectives of product purity and quantity. The sequence (maximum feed, no flow, maximum exhaust) is optimal. The experimental optimum is close to the calculated optimum. Dimensional analysis is used to determine parametric effects.

In the paper of Shendalman, et. al. (25), heatless adsorption, a cyclic gas adsorption process which employes pressure changes for bed regeneration, is experimentally investigated in the model system of CO_2 in excess He. Comparison is made with analytical results from a linear mathematical model of equilibrium adsorption of a trace component obtained by the method of characteristics. It is found that above a critical purge flow/feed flow ratio, the concentration of adsorbate in the product decreases with each increasing half-cycle from an initially saturated bed, while below the critical ratio an asymptotic adsorbate concentration is reached in agreement with theory. The theory predicts an exponential diminuation of CO_2 concentration with time. It is observed that the concentration levels decrease monotonically but not this rapidly. The model does provide considerable physical insight into the workings of heatless adsorption as well as a bound for process operation. The system CO_2-heluim on silica gel was studied using a two-bed system which was similar to Skarstrom's original system (37). Shendalman, et. al. (25) employed the equilibrium theory of Pigford, et. al. (7) to model heatless adsorption since this theory (7) was in good agreement with experimental results obtained for parametric pumping and heatless adsorption utilized the same basic principles of parametric pumping, the only complications being the repressurization and blowdown portions of the cycle. However, this was not true because where the theory (7) predicts a step decrease in concentration, the experimental results revealed a sawtooth pattern of concentration versus time. Though the local equilibrium theory does indicate the importance of the purge to feed ration, it seems that it is inappropriate to apply to heatless adsorption operation because of non-linear isotherms, rate processes, dispersion, and the intricate flow behavior during repressurization and blowdown.

Mitchell, et. al. (31) again used heatless adsorption and pressure changes on the model system of CO_2 in excess He except that they developed a non-equilibrium model in the attempt to model the repressurization and blowdown parts of the cycle. Transport in the pores was surmised to be the controlling mass transfer step, and they employed one lumped parameter expression to simulate the mass transfer. Isothermal column operation and axial dispersion were the assumptions adhered to again. Two assumptions that either the change of pressure was so fast that solid and gas concentrations remained constant or that the solid and gas were in equilibrium throughout the changes of pressure, were made for the repressurization and blowdown steps. The first assumption predicted more separation than was achieved, whereas the second assumption predicted considerably less separation than was experimentally discovered. A sawtooth pattern was predicted by the new theory when concentration was plotted versus time, however it did not predict totally the effect of the purge-to-feed ratio.

Systems Based on pH Difference

Sabadell, et. al. (12) utilized the recuperative mode to separate cations by means of a pH difference. As Sabadell, et. al. (12) explains, separations of aqueous solutions of Na^+ and K^+ have been obtained using a recuperative pH parametric pumping system. A chromatographic column of cation exchange resin was subjected simultaneously to an alternating axial displacement of solution and to an alternating pH gradient. The synchronous coupling of these two actions produced a concentration enrichment of 15-80% above the feed. Little separation of the two cations occurred for most of the reported runs, instead the cations were concentrated in the acidic reservoir. The ultimate separation factor found for $K^+ + Na^+$ was 1.84. The neutralization reaction that is undergone in the column provides the energy for the separation. Sabadell, et. al. (12) did not try to optimize the separation but did point out that theoretical calculations for pH parametric pumping will be more complex than for thermal parametric pumping.

EXPERIMENTAL

The laboratory scale equipment of both continuous and semicontinuous parametric pumps consisted of a jacketed glass column ninety centimeters in length and one centimeter in width which was packed with 50-100 mesh, calcium form analytical grade cation exchange resin (See Figure 1). The reservoirs at the two opposite ends of the column were two 50 cm^3 glass syringes operated by a dual infusion-withdrawal pump manufactured by Harvard Apparatus Company. A microswitch with stops was wired into the pump circuit to automatically reverse the action of the syringe plungers at the end of each half cycle.

The sources of hot and cold water supply were constant temperature hot and refrigerated baths. The baths were connected to the column and to recycle by solenoid valves wired to a dual timer so that hot water supply was always directed to the column jacket during upflow and cold water during downflow.

The feed was delivered to the top of the column by using a 50 cm^3 glass syringe as the feed reservoir and operating an infusion pump only. The product take-off valves were micrometer capillary valves used both to regulate flow and impose a small back pressure on the system.

Prior to each run the entire system, including the interstitial column volume, the bottom and top reservoirs, and the feed pump were filled with the feed mixture at ambient temperature. The reservoir syringes were set to deliver about 25 cm^3 per half cycle with a dead volume of 6.8 cm^3 in each syringe.

At the beginning of a continuous run the feed and reservoir pumps were started and the timer was activated. The bottom reservoir syringe pumped fluid into the bottom of the column and the timer simultaneously switched the solenoids to supply hot water (328 °K) to the jacket. At the end of the hot upflow half cycle, the microswitch on the pump automatically reversed the action of the reservoir syringes and the timer simultaneously switched the solenoids to supply cold water (278 °K) to the jacket. This procedure was repeated for each cycle during which the product take-off valves were opened and adjusted for the desired product flow rates. At the beginning of a semicontinuous run, the feed pump was shut off and the product take-off valves were closed during the hot upflow half cycle. However, during the cold downflow half cycle the column operates continuously as described previously.

For most runs the total cycle time used for this study was 2,400 seconds, that is 1,200 seconds of upflow followed by 1,200 seconds of downflow. (See Appendix, Table 4, p.61)

Method of Product Concentration Measurement and Calibration

Samples for analysis were taken from the product streams at the end of each cold half cycle for both modes of pump operation, and diluted for the purposes of measuring their respective concentrations. Depending on the run (see Table 1), 0.5 cm^3, 1 cm^3, or 2 cm^3 of bottom product was pipetted int 5.0 cm^3 of distilled water. The dilution factors for the bottom product samples were thus (5.0 + 0.5) divided by 0.5, (5.0 + 1.0) divided by 1.0, or (5.0 + 2.0) divided by 2.0 (i.e., the concentration of the bottom product was 11, 6, or 3.5 times that of the measured, diluted samples). Similarly, 1.0 cm^3 of top product was pipetted into 5.0 cm^3 of distilled water, resulting in a dilution factor of 6.

Note that for continuous runs 6 and 8, the bottom product analytical results were obtained from the bottom product samples collected during the hot half cycle. These hot half cycle analytical results were multiplied by the factor (1-b)/(1+b) to obtain the experimental values for the cold half cycle bottom product (See theory, p.41).

The product stream samples were analyzed by an automatic polarimeter. The polarimeter readings were made at ambient temperature. A calibration table of polarimeter readings corresponding to both fructose and glucose concentrations was made by measuring carefully (using pipettes) prepared solutions of known concentration (See Appendix, Table 3, p. 60). For runs involving the binary systems (glucose-water and fructose-water) the analysis was straightforward and the concentration of solute was linearly proportional to the polarimeter reading expressed in angular degree i.e., $R_g^o = \beta_g \eta_g$ for glucose and $R_f^o = \beta_f \eta_f$ for fructose (curves 1 and 2 of Figure 6). In the case of the ternary system, glucose-fructose-water, the analysis was somewhat complicated. The R^o for total sugars (glucose and fructose) was assumed to be that for glucose and for fructose, i.e.

$$R^o = R_g^o + R_f^o$$

$$R^o = \beta_f \eta + \eta_g (\beta_g - \beta_f) \qquad (1)$$

where

$$\eta = \eta_g + \eta_f$$

$$\beta_g = 9.614 \times 10^3$$

$$\beta_f = -16.796 \times 10^3$$

As shown in Equation 1, for a given η_g, a straight line results when R^o is plotted against η (curve 3 of Figure 6). Thus, knowing η_g one can determine η from R^o using Equation 1, and the fructose concentration η_f is obtained by subtraction of η_g from η. Note that η_g may be determined by the use of glucostat (Teller, (40)). However, for the present study η_g was found to be constant and equal to the feed concentration.

The feed solution was prepared using reagent grade glucose and fructose obtained from Fisher Scientific Co. The ternary runs required the aid of an analytical balance to help prepare their feed solutions since the accuracy of the glucose concentration was critical, it being a constant. Thus, another equation, which is a modification of equation 1, was developed to describe the concentration transients for both the top and bottom product streams. The initial or feed concentration, y_o, for the ternary runs was 1.423 x 10^{-4} gmoles/cm^3 for each respective sugar solute, i.e., for glucose, y_{og} = 1.423 x 10^{-4} gmoles/cm^3, and similarly for fructose, y_{of} = 1.423 x 10^{-4} gmoles/cm^3. Now, note again that for glucose $\eta_g = y_{og}$ throughout every ternary run. Therefore,

$$R^o = R^o_g + R^o_f$$

$$R^o = \beta_g \eta_g + \beta_f \eta_f$$

$$R^o/yo = \beta_g \eta_g/yo + \beta_f \eta_f/yo$$

$$\beta_f \eta_f/yo = R^o/yo - \beta_g \eta_g/yo$$

$$\eta_f/yo = 1 / \beta_f\, yo \, (R^o - \beta_g \eta_g)$$

and after substitution of the numerical values of yo, β_f, β_g

$$\eta_f/yo = \langle y_{BP2} \rangle nf/yo \text{ and } \langle y_{TP2} \rangle nf/yo = (1/-2.3901)(R^o - 1.3681) \qquad (2)$$

Thus the concentration measurement procedure involved obtaining bottom and top product samples, employing a dilution factor to all of the samples, then using polarimeter to obtain R^o, and then utilizing equation 2 to obtain the ratio of fructose concentration at any time to fructose and glucose feed concentration (concentration transients) for both bottom and top product streams. The experimental results for all runs are presented in tabular form in Table 4, p. 61 and in graphical form in Figures 7 and 8, pp. 78,79.

THEORY

Figures 3 and 4 show the continuous and the semicontinuous parametric pumping models considered here. Flow is upward during the hot half-cycle and downward during the cold half-cycle. Each half-cycle is $\frac{\pi}{\omega}$ time units in duration and the reservoir displacement volume is $Q(\frac{\pi}{\omega})$, where Q is the reservoir displacement rate. Each pump has dead or void volumes V_T and V_B for the top and bottom reservoirs respectively. The feed is directed to the top of the columns at the flow rate $(\phi_T + \phi_B)Q$. The top product flow rate is $\phi_T Q$ and the bottom product flow rate is $\phi_B Q$, where ϕ_T and ϕ_B are ratios of the top and bottom product rates to the reservoir displacement rate.

For processes inside the column, Pigford's, et. al. (7) assumption that local interphase equilibrium exists with a linear distribution law having a temperature dependent distribution coefficient, is employed here. Also there is negligible axial diffusion, temperature changes between hot and cold cycles are instantaneous, plug flow exists, and the fluid density is constant. A further assumption that the multicomponent mixture may be treated as n pairs of pseudo binary systems. Each system includes one solute and the common inert solvent and could be characterized by a dimensionless equilibrium parameter b_i and corresponding values of the penetration distances of the hot and cold cycles L_{1i} and L_{2i}. L_{1i} and L_{2i} can be expressed in terms of ϕ_B and the equilibrium parameter b_i. For the continuous pump

$$L_{1i} = \frac{Q(1 - \phi_B)}{A\,\varepsilon\,(1 + m_o)\ (1 - b_i)}\ \frac{\pi}{\omega} \tag{3}$$

$$L_{2i} = \frac{Q(1 + \phi_B)}{A\,\varepsilon\,(1 + m_o)\ (1 + b_i)}\ \frac{\pi}{\omega} \tag{4}$$

For the case of the semicontinuous pump

$$L_{1i} = \frac{Q}{A\,\varepsilon(1 + m_o)\ (1 - b_i)}\ \frac{\pi}{\omega} \tag{5}$$

and

$$L_{2i} = \frac{Q(1 + \phi_B)}{A\,\varepsilon\,(1 + m_o)\ (1 + b_i)}\ \frac{\pi}{\omega} \tag{6}$$

where A is the area normal to flow, ε is the fraction of the column occupied by void (fluid) volume, and m_o is another equilibrium parameter which like b_i is only a function of hot and cold temperatures. Actually, b_i is a measure of the extent of movement of solute between phases as a result of a change in column temperature and may be as small as zero (as it is for glucose) in which the equilibrium distribution is insensitive to temperature or as large as unity for which the equilibrium distribution is very sensitive to temperature. The equilibrium parameter m_o is a quantity proportional to the thermal equilibrium constant at the mean temperature of the cycle.

The pump performance depends on the relative magnitudes of L_{1i}/L_{2i} and the height of the column h. There are three possible regions of pump operations depending on L_{1i}/L_{2i} and h,

Region 1, $\frac{L_{1i}}{L_{2i}} \geq 1$ (or $\phi_B < b_i$ for the continuous pump) (7)
(or $\phi_B < \frac{2b_i}{1 - b_i}$ for the semicontinuous pump and $L_{2i} \leq h$

Region 2, $\frac{L_{1i}}{L_{2i}} < 1$ (or $\phi_B > b_i$ for the continuous pump)
(or $\phi_B > \frac{2b_i}{1 + b_i}$ for the semicontinuous
pump, and $L_{1i} \leq h$

Region 3, L_{1i} and $L_{2i} > h$

By treating the multicomponent mixture as a series of pseudo binary systems, the multicomponent separation could be predicted by the existing mathematical expressions for binary systems (Chen, et. al. (15, 22, 27)). Their derivations are not repeated here but note that it has been found that at steady state ($n \rightarrow \infty$) solute removal from the bottom product stream $\phi_B Q$ is complete in Region 1 and only partial in Regions 2 and 3.

Now consider a mixture containing s solutes, each with its own b_i and

$$b_i > b_2 > \text{---}\ b_k \geq \phi_B > b_k + 1 \text{---} > b_s \tag{8}$$

where subscripts 1, 2, etc., refer to solutes 1, 2, etc. Furthermore

$$\begin{aligned} L_{2i} &\leq h \quad \text{when} \quad i = 1, 2, \text{- - -}\ k \\ L_{1i} &\leq h \quad \text{when} \quad i = k\ \ 1, \text{- - -}\ s \end{aligned} \tag{9}$$

At steady state the components, i = 1, 2, --- k for which the operations are indicated in Region 1, would appear only in the top product stream, and the remaining components (k + 1, --- s) would appear in both the top and bottom product streams. In the extreme case where k = s the bottom product stream would consist only of pure solvent By proper adjustment of ϕ_B in Equation 8 a solute split could be made which is analogous to that obtained by a multicomponent distillation column.

RESULTS AND DISCUSSION

The experimental parameters are shown in Table 4 and the data are plotted in Figures 7 to 9. The equations previously derived (Chen, et. al., (22) (27)) were used to calculate the concentration transients, and computed results corresponding to the experimental runs are also plotted in Figures 7 to 9. These results compare reasonably well with the observed values for both continuous and semicontinuous pump operations. For the continuous pump the feed and product streams flow steadily both in upflow and downflow cycles, while the semicontinuous pump is operated batch-wise during upflow and continuously during downflow.

Figure 7 illustrates concentration transients for both glucose and fructose in water. For glucose $\langle y_{TP2} \rangle_n / y_o$ and $\langle y_{BP2} \rangle_n / y_o$ are essentially independent of n. This means that the separation of glucose from water does not occur to any appreciable extent, and the concentrations of top and bottom product streams may be considered to be constant and equal to the feed concentration. In other words the equilibrium distribution between glucose and the resin is insensitive to temperature and the equilibrium parameter, $b_g = 0$ (Chen and Hill, (15)). For the case of fructose-water, $\langle y_{BP2} \rangle_n / y_o$ decreases as n increases and as the theory predicts, approaches zero as n becomes large. As $n \rightarrow \infty$, fructose removal from the bottom product stream can be complete and the top product stream must carry away all of the fructose supplied by the feed stream and reach a steady state value, $\langle y_{TP2} \rangle_{\infty} / y_o$. Consequently, b_f can be obtained by a method described by Chen et. al. (22) and was found to be 0.058. Note that b_i is a function of hot and cold temperatures only.

Figure 8 shows the concentration transients for fructose in ternary systems (glucose - fructose - water). In the computations it is assumed that the system contains two pseudobinaries, each binary consisting of one solute as one component and the common solvent as the other component (i.e., glucose - water and fructose - water). Each system could be characterized by a dimensionless equilibrium parameter b_i and corresponding values of L_{1i} and L_{2i} (Chen et. al., (34)). One can see that for all cases $\langle y_{BP2} \rangle_n / y_o$ decreases as n increases. The slope α (of Log $\langle y_{BP2} \rangle_n / y_o$ versus n) is coincident with that in the binary mixture (fructose - water, Figure 7), since α depends only on the values of C_2 and b_i where C_2 is defined as the ratio of dead volume of the bottom reservoir to the displacement.

Chen and Hill, (15) have shown that there are three possible regions of pump operations. Regions 1, 2, and 3, depending on the relative magnitudes of penetration distances L_{1i} and L_{2i}, and the height of the column, h. As long as,

$$b_g < \phi_B \leq b_f \text{ and } L_{2f} \leq h \qquad (10)$$

for the continuous pump, and

$$\left(\frac{2b}{1 - b}\right)_g < \phi_B \leq \left(\frac{2b}{1 - b}\right)_f \quad \text{and } L_{2f} \leq h \qquad (11)$$

for the semicontinuous pump, the operation is in Region 1 for fructose and Region 2 for glucose. At steady state ($n \to \infty$) fructose appears only in the top product stream, but glucose would appear in both top and bottom product streams. However, as stated before for the present study $b_g = 0$, and the glucose concentration in either top or bottom product streams is identical to that of the feed.

It should be emphasized that the performance characteristics of both continuous and semicontinuous pumps are similar in nature. The main difference between the two pumps is the difference in the loci of switching points between Regions 1 and 2 (Chen and Hill, (15)). For the continuous pump, the switching points correspond to the condition $\phi_B = b_i$. In the case of the semicontinuous pump, the condition is $\phi_B = 2b_i/(1 - b_i)$.

Figure 9 shows the effects of ϕ_T and ϕ_B on the degree of enrichment in the top product stream. ϕ_T and ϕ_B are defined as the ratios of the top and bottom product flow rates to the reservoir displacement rate. Provided that the pumps are operated in Region 1 (see Eqs. 10 and 11), the top product concentration at $n \to \infty$ would be $\langle y_{TP2} \rangle_\infty / y_o = 1 + \phi_B/\phi_T$. As ϕ_B increases, $\langle y_{TP2} \rangle_\infty / y_o$ increases. Furthermore, for a given value of ϕ_B, by adjustment of ϕ_T to an arbitrarily low value one may obtain an arbitrarily high value of $\langle y_{TP2} \rangle_\infty / y_o$.

This last point is graphically illustrated via theoretical results in Figs. 10 and 12, where the number of cycles of pump operation (n) is extended to 100 cycles. Note that when ϕ_B and b_i are increased (Figure 12), increasingly higher degrees of enrichment in the top product stream are obtained.

Figures 11 and 13 represent theoretical separation factors for the glucose-fructose-water system again for 100 cycles of pump operation. They take the form of the graphs of the concentration transients, because they are mathematically exactly that. Expressed mathematically, the separation factors for the bottom and the top product streams are defined as $\langle y_{BP2}\rangle_f / \langle y_{BP2}\rangle_g$ and $\langle y_{TP2}\rangle_f / \langle y_{TP2}\rangle_g$. Now, taking the bottom product stream to serve as an example for

$$(\langle y_{BP2}\rangle / y_o)_f = x_f \quad \text{and} \tag{12}$$

$$(\langle y_{BP2}\rangle / y_o)_g = x_g \tag{13}$$

$$\frac{\langle y_{BP2}\rangle_f}{\langle y_{BP2}\rangle_g} = \frac{x_f \ y_{of}}{x_g \ y_{og}} \tag{14}$$

however, $y_{of} = y_{og} = 1.423 \times 10^{-4}$ gmoles/cm^3

and $x_g = 1$ (See Appendix, Fig. 7, p. 78)

Therefore

$$\frac{\langle y_{BP2}\rangle_f}{\langle y_{BP2}\rangle_g} = x_f = \frac{\langle y_{BP2}\rangle_{n \ f}}{y_{of}} \tag{15}$$

Similarly for the top product stream

$$\frac{\langle y_{TP2}\rangle_f}{\langle y_{TP2}\rangle_g} = t_f = \frac{\langle y_{TP2}\rangle_{n \ f}}{y_{of}} \tag{16}$$

This is why the ordinates are labeled the way they are in Figures 11 and 13. Again it is shown that when ϕ_B and b_i increase (See Figure 13),

$\langle y_{TP2} \rangle_n / y_o$ increases accordingly and $\langle y_{BP2} \rangle_n / y_o$ decreases accordingly, provided that the pumps are operated in Region 1 (See Equations 10 and 11).

CONCLUSIONS AND SIGNIFICANCE

A simple means of predicting multicomponent separations in both continuous and semicontinuous thermal parametric pumping is presented. A multicomponent system (glucose-fructose-water) is treated as a series of pseudo binary systems. Each binary system consists of one of the solutes as one component and the common solvent as the other component. This approach permits the use of existing transient and steady state equations for binary systems (Chen and Hill, (15); Chen, et. al., (22)). Experimental data for the concentration transients agree reasonably well with the analytical predictions.

It is shown that the thermal parametric pump is capable of separating components in a multicomponent mixture (in this case, separating isomers) and, as a theoretical limit, of attaining infinite separation factors. Also, the net movement of concentration fronts through the adsorption column is found to be important in determining the pump performance. Those solutes for which the net movement is upward would, at steady state, appear only in the top product. The remaining solute(s) would appear in both the top and bottom products. In the limiting case, it is possible for the bottom product to consist solely of pure solvent. This would result when all solutes in a mixture are very strongly adsorbed or desorbed in a given cycle or when the flow rate of the bottom product is very small.

For the glucose-fructose-water model system, the optimal pump performance would probably result from the use of a semicontinuous pump operating with a maximum displacement rate, the minimum cycle time, a greater amount of bottom product collected with no corresponding change in feed delivery, and a maximum possible difference in hot and cold temperature.

As the experimental results reveal, the glucose-fructose-water system was an equilibrium system. Also, it can again be concluded that systems having relatively smaller values of b_i will have much better steady state separations for a given cycle time.

NOMENCLATURE

b = equilibrium parameter, dimensionless
C_1 = top reservoir dead volume/displacement, dimensionless
C_2 = bottom reservoir dead volume/displacement, dimensionless
h = column height, m
L_1 = penetration distance for hot half-cycle, m
L_2 = penetration distance for cold half-cycle, m
n = number of cycles of pump operation
Q = reservoir displacement rate, cm^3/s
R^o = polarimeter reading, angular degree
T_1 = column temperature during upflow, ^oK
T_2 = column temperature during downflow, ^oK
y_o = initial or feed concentration of solute, g moles/cm^3
$\langle y_{BP2} \rangle n$ = average bottom product concentration of solute during downflow at n^{th} cycle, g moles/cm^3
$\langle y_{TP2} \rangle n$ = averate top product concentration of solute during downflow at n^{th} cycle, g moles/cm^3
η = concentration, g moles/cm^3
β = R^o / η
ϕ_B = bottom product volumetric flow rate over reservoir displacement rate, dimensionless
ϕ_T = top product volumetric flow rate over reservoir displacement rate, dimensionless
$\frac{\pi}{\omega}$ = duration of half cycle, s

SUBCRIPTS

f = fructose

g = glucose

i = solute i

∞ = steady state

APPENDIX

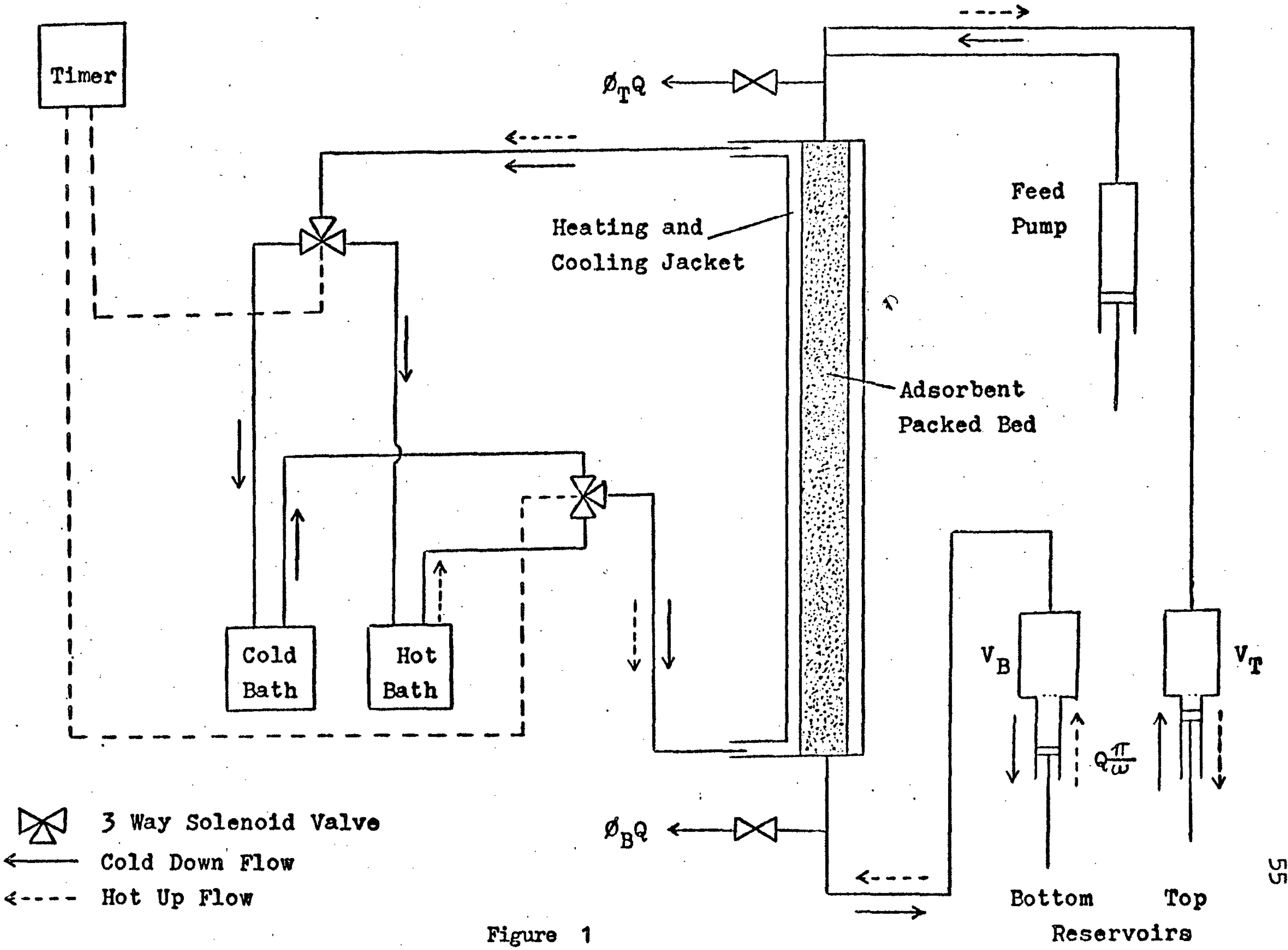
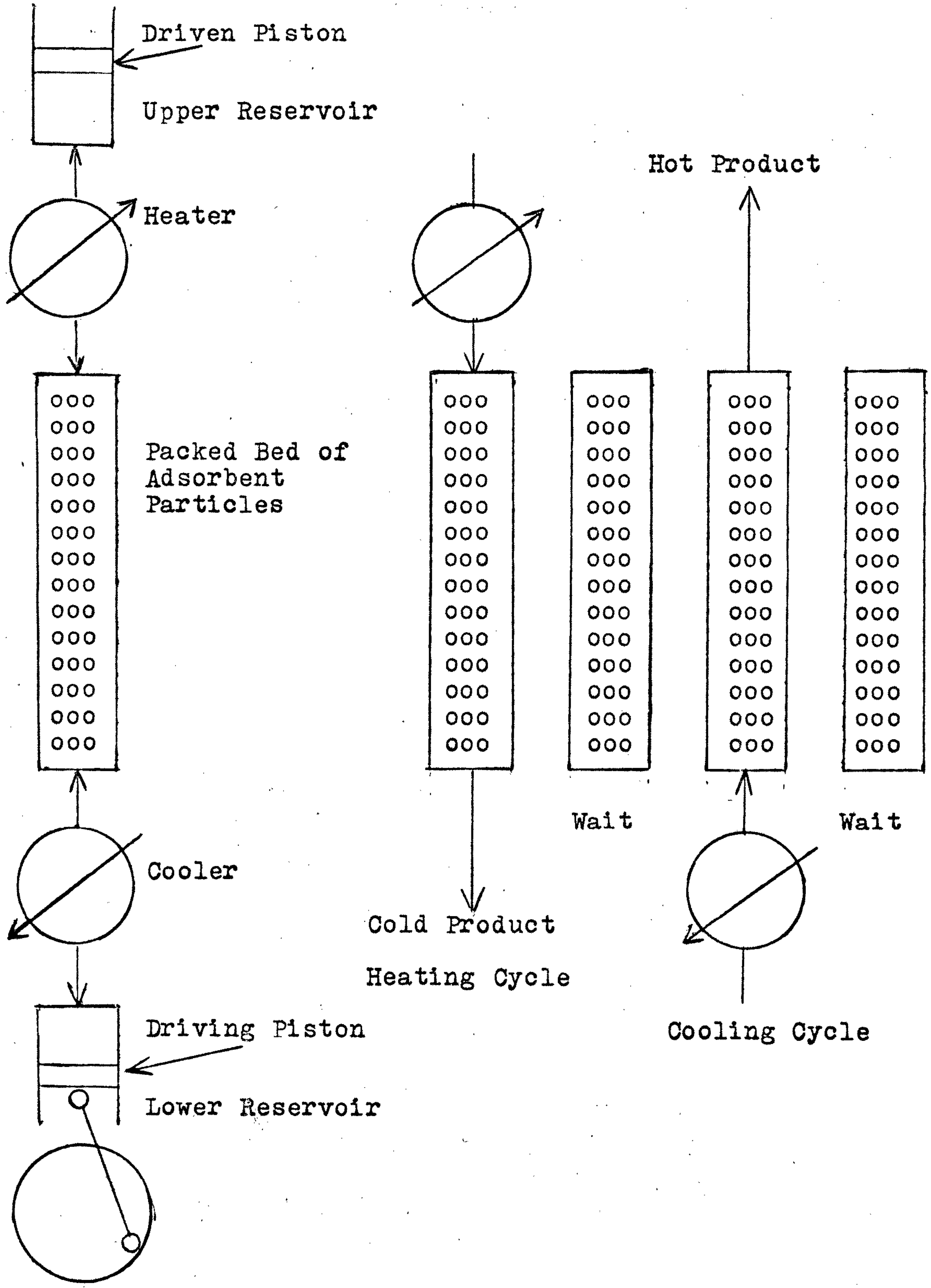

Timer
$\phi_T Q$
Heating and Cooling Jacket
Feed Pump
Adsorbent Packed Bed
Cold Bath
Hot Bath
V_B
V_T
$Q\frac{\pi}{\omega}$
$\phi_B Q$
Bottom
Top
Reservoirs
3 Way Solenoid Valve
Cold Down Flow
Hot Up Flow

Figure 1

FIGURE 2



Experimental Arrangements for Closed System Recuperative Operation

Operation for an Open Recuperative System with Trapezoidal Displacement

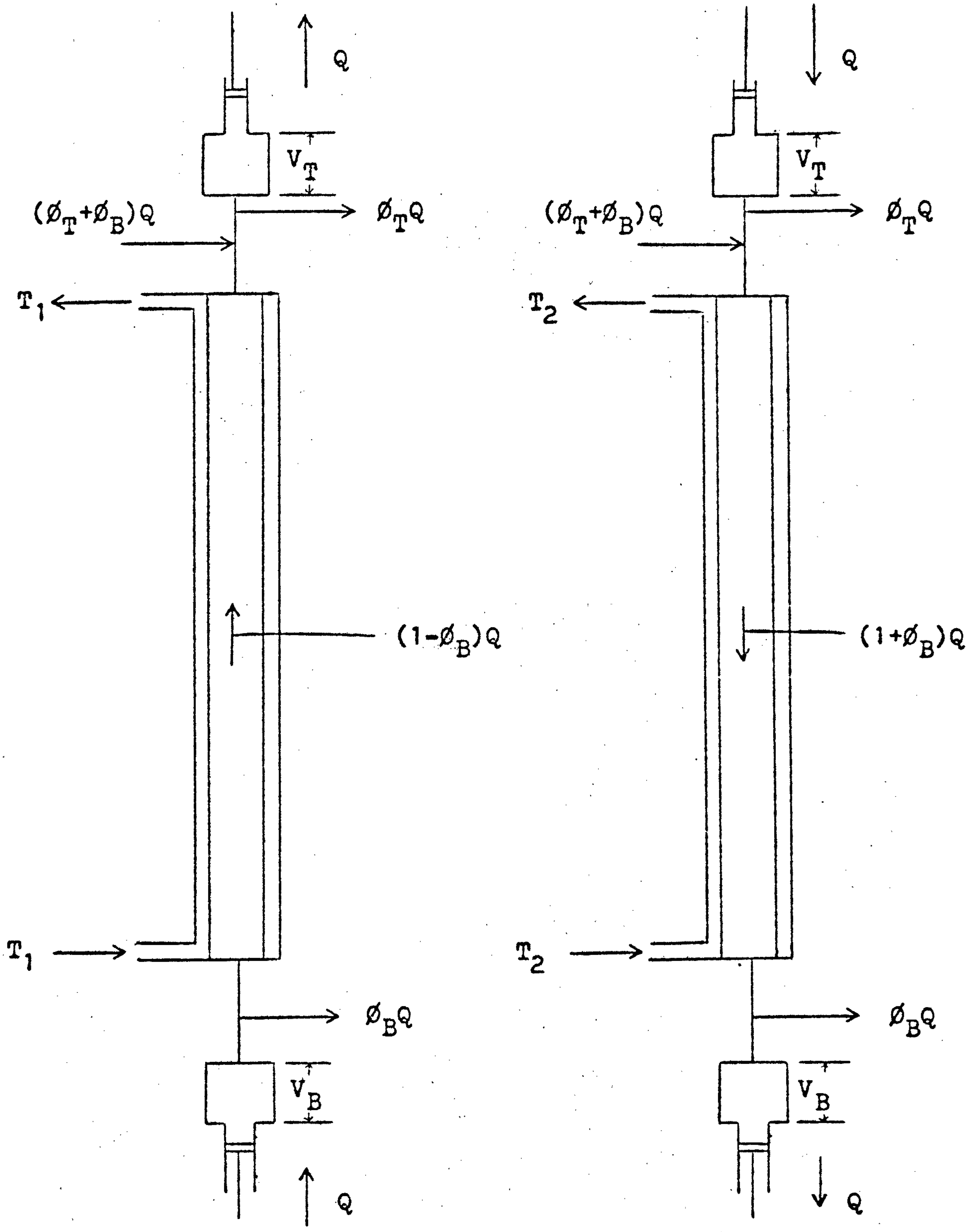


Figure 3 — The Continuous Parametric Pump

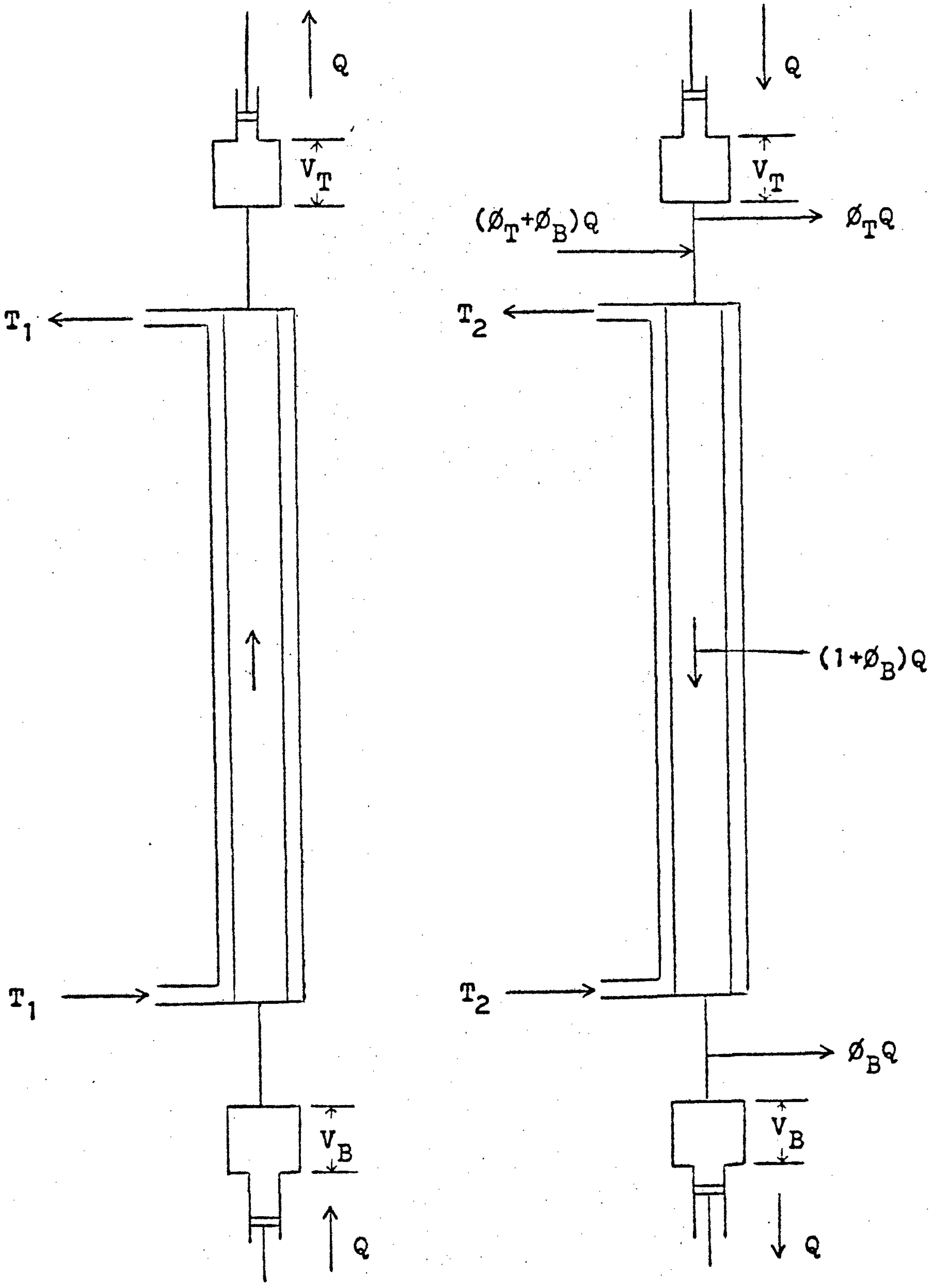


Figure 4 — The Semicontinuous Parametric Pump

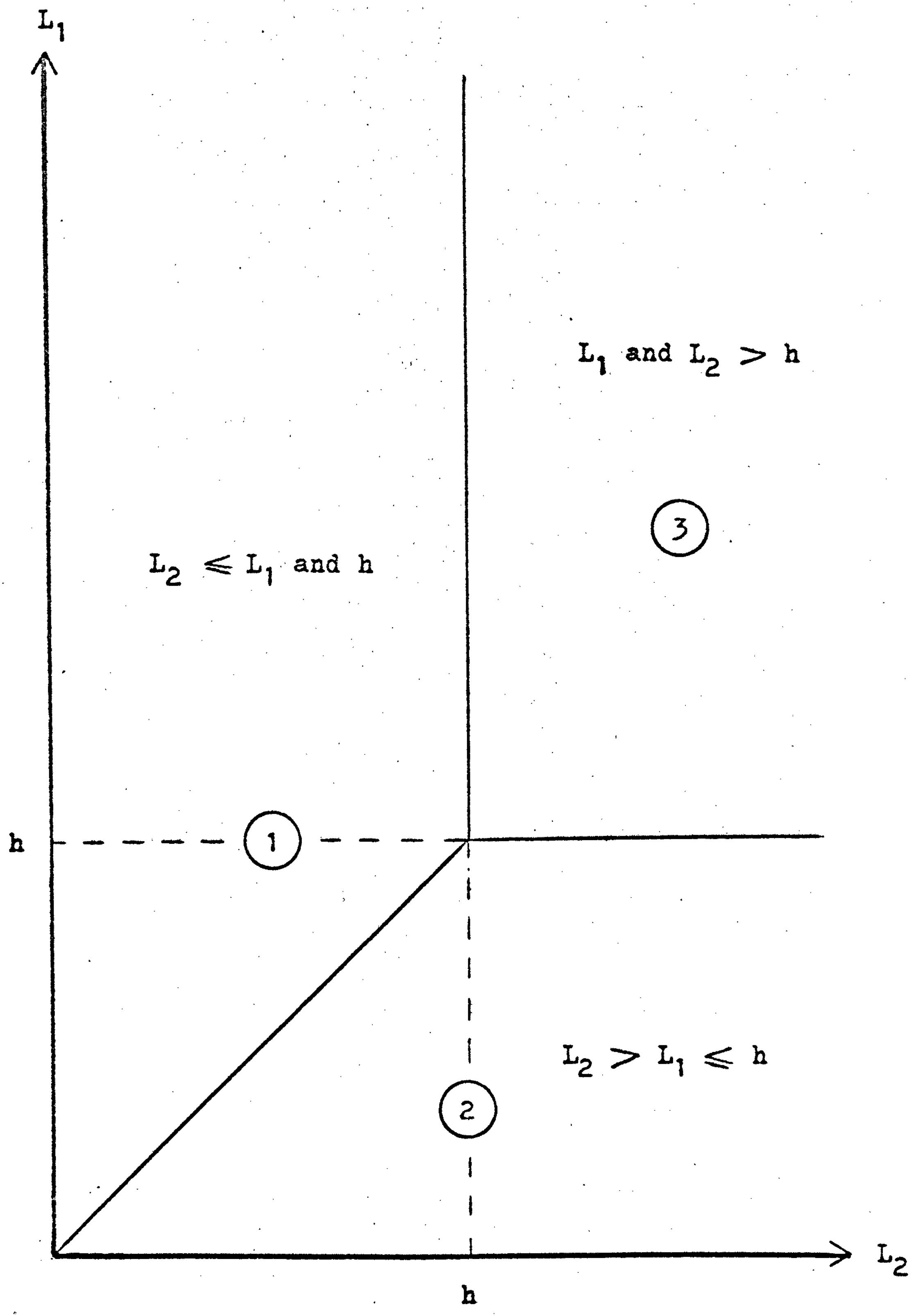


Figure 5 — The Three Regions of Parametric Pump Operation

TABLE 3

Calibration Data

Fructose Concentration $(\text{gmoles/cm}^3) \times 10^6$	Net Polarimeter Reading (Angular Degree)
1110.1	-18.670
555.1	- 9.179
277.5	- 4.557
138.8	- 2.311
69.4	- 1.151
34.7	- 0.577
2.17	- 0.028

Glucose Concentration $(\text{gmoles/cm}^3) \times 10^6$	Net Polarimeter Reading (Angular Degree)
1110.1	+10.943
555.1	+ 5.238
277.5	+ 2.640
138.8	+ 1.338
2.17	+ 0.030

TABLE 4 - EXPERIMENTAL AND MODEL PARAMETERS

$\frac{\pi}{\omega}^{*} = 1{,}200$ s., $T_1 = 328^{o}K$, $T_2 = 278^{o}K$, $b_g = 0$, $b_f = 0.058$

$h = 0.9$ m, $C_1 = 0.272$, $C_2 = 0.272$, $Q\left(\frac{\pi}{\omega}\right) = 25$ cm^3

$y_{og}^{**} = 1.423 \times 10^{-4}$ g moles/cm^3

$y_{of}^{***} = 1.423 \times 10^{-4}$ g moles/cm^3

			ϕ_B	$\phi_T+\phi_B$	L_{1f}(m)	L_{2f}(m)	$\frac{\langle y_{TP2}\rangle_\infty}{y_o}$
1.	Glucose-Water	Semicontinuous	0.032	0.40	-	-	-
2.	Fructose-Water	Semicontinuous	0.032	0.40	0.633	0.582	1.09
3.	Glucose-Fructose-Water	Semicontinuous	0.032	0.20	0.633	0.582	1.19
4.	Glucose-Fructose-Water	Semicontinuous	0.060	0.28	0.636	0.600	1.27
5.	Glucose-Fructose-Water	Continuous	0.030	0.28	0.614	0.580	1.12
6.	Glucose-Fructose-Water	Continuous	0.056	0.28	0.596	0.593	1.25
7.	Glucose-Fructose-Water	Semicontinuous	0.080	0.20	0.633	0.609	1.67
8.	Glucose-Fructose-Water	Continuous	0.056	0.20	0.600	0.598	1.39

---

* Except Run 6 for which $\frac{\pi}{\omega} = 1{,}800$ s.

** For Run 1, $y_{og} = 2.774 \times 10^{-4}$ g moles/cm^3

*** For Run 2, $y_{of} = 2.601 \times 10^{-4}$ g moles/cm^3

TABLE 5

Experimental Results of Run # 1

L_1 = - L_2 = - Region Two

	$R^{o}BP$ (diluted)	$R^{o}TP$ (diluted)	$\langle y_{BP} \rangle n \times 10^6$ (diluted)	$\langle y_{TP} \rangle n \times 10^6$ (diluted)	$\langle y_{BP} \rangle n/yo$	$\langle y_{TP} \rangle n/yo$
1.	-	-	-	-	-	-
2.	-	-	-	-	-	-
3.	+.251	-	26.06	-	1.0334	-
4.	+.250	+.417	25.95	43.29	1.0291	0.9364
5.	+.248	-	25.75	-	1.0211	-
6.	+.260	+.433	26.99	44.95	1.0703	0.9723
7.	+.248	-	25.75	-	1.0211	-
8.	+.243	+.451	25.23	46.82	1.0005	1.0127
9.	+.241	+.446	25.02	46.30	0.9922	1.0015
10.	+.232	+.450	24.08	46.72	0.9549	1.0106
11.	-	-	-	-	-	-
12.	-	-	-	-	-	-
13.	-	-	-	-	-	-
14.	-	-	-	-	-	-
15.	-	-	-	-	-	-
16.	-	-	-	-	-	-

TABLE 6

Experimental Results of Run # 2

L_1 = 63.34 L_2 = 58.18 Region One

	R^oBP (diluted)	R^oTP (diluted)	$\langle y_{BP}\rangle n \times 10^6$ (diluted)	$\langle y_{TP}\rangle n \times 10^6$ (diluted)	$\langle y_{BP}\rangle n/yo$	$\langle y_{TP}\rangle n/yo$
1.	-.396	-	23.62	-	0.9990	-
2.	-.389	-	23.20	-	0.9812	-
3.	-.349	-	20.81	-	0.8802	-
4.	-.330	-	19.68	-	0.8324	-
5.	-.282	-	16.82	-	0.7114	-
6.	-.261	-.933	15.57	55.64	0.6585	1.2836
7.	-	-	-	-	-	-
8.	-.189	-	11.27	-	0.4767	-
9.	-.181	-	10.79	-	0.4564	-
10.	-.159	-1.047	9.48	62.44	0.4010	1.4405
11.	-.150	-	8.95	-	0.3785	-
12.	-.134	-1.038	7.99	61.90	0.3379	1.4280
13.	-.115	-	6.86	-	0.2901	-
14.	-.115	-1.005	6.86	59.94	0.2901	1.3828
15.	-.110	-.991	6.56	59.10	0.2775	1.3634
16.	-.098	-1.046	5.84	62.38	0.2470	1.4391

```
***************
H=     90.000      B=      0.058
(PHO)UPPER=      0.368       (PHO)LOWER=       0.032
DIST1=    63.336      DIST2=     58.184
(YT/YO)INF=           0.10870E 01       (YB/YO)INF=          0.00000E 00
C1=                  0.272             C2=                 0.272
(PHO)LOWER/FEED=      0.080      FEED=      0.400

     N              YT/YO                      YB/YO                        SF

     1           0.10000E 01              0.89018E 00               0.11234E 01
     2           0.10891E 01              0.81333E 00               0.13391E 01
     4           0.11974E 01              0.67895E 00               0.17636E 01
     6           0.12521E 01              0.56677E 00               0.22092E 01
     8           0.12752E 01              0.47313E 00               0.26953E 01
    10           0.12702E 01              0.39496E 00               0.32160E 01
    12           0.12531E 01              0.32971E 00               0.38006E 01
    14           0.12323E 01              0.27523E 00               0.44772E 01
    16           0.12116E 01              0.22976E 00               0.52735E 01
    18           0.11927E 01              0.19180E 00               0.62186E 01
    20           0.11761E 01              0.16011E 00               0.73456E 01
    22           0.11618E 01              0.13366E 00               0.86925E 01
    24           0.11497E 01              0.11157E 00               0.10304E 02
    26           0.11394E 01              0.93141E-01               0.12233E 02
    28           0.11308E 01              0.77752E-01               0.14544E 02
    30           0.11236E 01              0.64906E-01               0.17311E 02
    32           0.11175E 01              0.54182E-01               0.20626E 02
    34           0.11125E 01              0.45230E-01               0.24596E 02
    36           0.11083E 01              0.37758E-01               0.29353E 02
    38           0.11048E 01              0.31519E-01               0.35050E 02
    40           0.11018E 01              0.26312E-01               0.41875E 02
```

TABLE 7

Experimental Results of Run # 3

L_1 = 63.34 L_2 = 58.18   Region One

	R^o_{BP} (diluted)	R^o_{TP} (diluted)	$\langle y_{BP}\rangle n/yo$	$\langle y_{TP}\rangle n/yo$
1.	-	-	-	-
2.	-.036	-	0.738	-
3.	-	-	-	-
4.	-.002	-.176	0.582	1.01
5.	-	-	-	-
6.	+.023	-.241	0.467	1.18
7.	-	-	-	-
8.	+.031	-	0.430	-
9.	+.049	-	0.347	-
10.	+.047	-.320	0.356	1.38
11.	+.058	-	0.305	-
12.	-	-.334	-	1.41
13.	+.065	-	0.273	-
14.	+.071	-.352	0.246	1.46
15.	+.073	-	0.236	-
16.	+.079	-.349	0.209	1.45
17.	+.080	-	0.204	-
18.	+.085	-	0.181	-

```
***************
 H=     90.000      B=      0.058
 (PHO)UPPER=      0.168      (PHO)LOWER=      0.032
 DIST1=     63.336      DIST2=     58.184
 (YT/YO)INF=           0.11905E 01        (YB/YO)INF=          0.00000E 00
 C1=                 0.272             C2=                  0.272
 (PHO)LOWER/FEED=      0.160       FEED=      0.200


       N                YT/YO                     YB/YO                     SF

       1             0.10000E 01               0.89018E 00               0.11234E 01
       2             0.10891E 01               0.81333E 00               0.13391E 01
       4             0.12335E 01               0.67895E 00               0.18168E 01
       6             0.13424E 01               0.56677E 00               0.23684E 01
       8             0.14198E 01               0.47313E 00               0.30008E 01
      10             0.14599E 01               0.39496E 00               0.36963E 01
      12             0.14741E 01               0.32971E 00               0.44710E 01
      14             0.14715E 01               0.27523E 00               0.53464E 01
      16             0.14584E 01               0.22976E 00               0.63476E 01
      18             0.14393E 01               0.19180E 00               0.75040E 01
      20             0.14171E 01               0.16011E 00               0.88506E 01
      22             0.13939E 01               0.13366E 00               0.10429E 02
      24             0.13710E 01               0.11157E 00               0.12288E 02
      26             0.13493E 01               0.93141E-01               0.14486E 02
      28             0.13291E 01               0.77752E-01               0.17094E 02
      30             0.13108E 01               0.64906E-01               0.20195E 02
      32             0.12944E 01               0.54182E-01               0.23889E 02
      34             0.12798E 01               0.45230E-01               0.28295E 02
      36             0.12670E 01               0.37758E-01               0.33557E 02
      38             0.12558E 01               0.31519E-01               0.39844E 02
      40             0.12462E 01               0.26312E-01               0.47361E 02
```

TABLE 8

Experimental Results of Run # 4

$L_1 = 63.59$ $L_2 = 60.00$ Region One

	R^o_{BP} (diluted)	R^o_{TP} (diluted)	$\langle y_{BP} \rangle$ n/yo	$\langle y_{TP} \rangle$ n/yo
1.	-	-	-	-
2.	-	-	-	-
3.	-	-	-	-
4.	+.012	-.153	0.542	0.956
5.	-	-	-	-
6.	+.053	-.221	0.439	1.13
7.	-	-	-	-
8.	+.081	-	0.369	-
9.	-	-	-	-
10.	+.111	-.306	0.294	1.34
11.	-	-	-	-
12.	+.105	-.342	0.309	1.43
13.	-	-	-	-
14.	+.123	-.327	0.264	1.39
15.	-	-	-	-
16.	+.127	-.330	0.254	1.40
17.	-	-	-	-
18.	+.143	-	0.213	-
19.	-	-	-	-
20.	+.148	-	0.201	-

```
***************
H=     90.000      B=      0.058
(PHO)UPPER=        0.220       (PHO)LOWER=        0.060
DIST1=    63.587      DIST2=     60.000
(YT/YC)INF=             0.12727E 01      (YB/YC)INF=            0.00000E 00
C1=                    0.272              C2=                      0.272
(PHO)LOWER/FEED=      0.214     FEED=      0.280

     N               YT/YO                        YB/YO                          SF

     1            0.10000E 01                0.89018E 00                0.11234E 01
     2            0.10915E 01                0.81333E 00                0.13420E 01
     4            0.12298E 01                0.67895E 00                0.18113E 01
     6            0.13244E 01                0.56677E 00                0.23367E 01
     8            0.13891E 01                0.47313E 00                0.29359E 01
    10            0.14309E 01                0.39496E 00                0.36229E 01
    12            0.14476E 01                0.32971E 00                0.43905E 01
    14            0.14480E 01                0.27523E 00                0.52609E 01
    16            0.14391E 01                0.22976E 00                0.62633E 01
    18            0.14253E 01                0.19180E 00                0.74311E 01
    20            0.14094E 01                0.16011E 00                0.88029E 01
    22            0.13933E 01                0.13366E 00                0.10424E 02
    24            0.13777E 01                0.11157E 00                0.12348E 02
    26            0.13634E 01                0.93141E-01                0.14638E 02
    28            0.13505E 01                0.77752E-01                0.17369E 02
    30            0.13390E 01                0.64906E-01                0.20630E 02
    32            0.13290E 01                0.54182E-01                0.24529E 02
    34            0.13204E 01                0.45230E-01                0.29193E 02
    36            0.13130E 01                0.37758E-01                0.34774E 02
    38            0.13066E 01                0.31519E-01                0.41455E 02
    40            0.13012E 01                0.26312E-01                0.49455E 02
```

TABLE 9

Experimental Results of Run # 5

$L_1 = 61.38$ $L_2 = 58.02$ Region One

	R^o_{BP} (diluted)	R^o_{TP} (diluted)	$\langle y_{BP} \rangle$ n/yo	$\langle y_{TP} \rangle$ n/yo
1.	-	-	-	-
2.	-	-	-	-
3.	-	-	-	-
4.	+.010	-.210	0.547	1.10
5.	-	-	-	-
6.	+.039	-.227	0.474	1.14
7.	-	-	-	-
8.	+.073	-	0.389	-
9.	-	-	-	-
10.	+.077	-.239	0.379	-
11.	-	-	-	-
12.	+.080	-.241	0.372	1.18
13.	-	-	-	-
14.	+.052	-.229	-	1.15
15.	-	-	-	-
16.	+.052	-.250	-	1.20

```
***************
 H=    90.000      B=      0.058
 (PHO)UPPER=       0.250       (PHO)LOWER=      0.030
 DIST1=    61.377      DIST2=     58.015
 (YT/YO)INF=            0.11199E 01       (YB/YC)INF=             0.00000E 00
 C1=                  0.272             C2=                      0.272
 (PHO)LOWER/FEED=       0.107      FEED=      0.280

        N                 YT/YO                         YB/YO                        SF

        1              0.10000E 01                  0.89018E 00                 0.11234E 01
        2              0.10711E 01                  0.81333E 00                 0.13170E 01
        4              0.11533E 01                  0.67895E 00                 0.16987E 01
        6              0.11920E 01                  0.56677E 00                 0.21031E 01
        8              0.12101E 01                  0.47313E 00                 0.25577E 01
       10              0.12186E 01                  0.39496E 00                 0.30855E 01
       12              0.12175E 01                  0.32971E 00                 0.36927E 01
       14              0.12085E 01                  0.27523E 00                 0.43909E 01
       16              0.11973E 01                  0.22976E 00                 0.52109E 01
       18              0.11861E 01                  0.19180E 00                 0.61838E 01
       20              0.11759E 01                  0.16011E 00                 0.73441E 01
       22              0.11670E 01                  0.13366E 00                 0.87311E 01
       24              0.11594E 01                  0.11157E 00                 0.10391E 02
       26              0.11529E 01                  0.93141E-01                 0.12378E 02
       28              0.11475E 01                  0.77752E-01                 0.14759E 02
       30              0.11430E 01                  0.64906E-01                 0.17610E 02
       32              0.11392E 01                  0.54182E-01                 0.21025E 02
       34              0.11360E 01                  0.45230E-01                 0.25116E 02
       36              0.11334E 01                  0.37758E-01                 0.30017E 02
       38              0.11311E 01                  0.31519E-01                 0.35887E 02
       40              0.11293E 01                  0.26312E-01                 0.42920E 02
```

TABLE 10

Experimental Results of Run # 6

$L_1 = 59.57$ $L_2 = 59.32$ Region One

	$R^o{}_{BP}$ (diluted)	$R^o{}_{TP}$ (diluted)	$\langle y_{BP}\rangle^* n/yo$	$\langle y_{TP}\rangle n/yo$
1.	-	-	-	-
2.	-	-	-	-
3.	-	-	-	-
4.	-.084	-.232	0.619	1.15
5.	-	-	-	-
6.	+.015	-.258	0.490	1.22
7.	-	-	-	-
8.	+.074	-.273	0.413	1.26
9.	-	-	-	-
10.	+.069	-.291	0.419	1.30
11.	-	-	-	-
12.	+.058	-.305	0.380	1.34
13.	-	-	-	-
14.	+.067	-.299	0.360	1.32
15.	-	-	-	-
16.	+.087	-.289	0.315	1.30

$$*\frac{\langle y_{BP_2}\rangle n}{yo} = \left(\frac{1-b}{1+b}\right) \frac{\langle y_{BP_1}\rangle n}{yo}$$

```
****************
 H=     90,000      B=      0.058
 (PHO)UPPER=        0.224       (PHO)LOWER=        0.056
 DIST1=    59.567      DIST2=    59,316
 (YT/YO)INF=             0.12500E 01      (YB/YO)INF=              0.00000E 00
 C1=                   0.272            C2=                      0.272
 (PHO)LOWER/FEED=      0.200       FEED=      0,280

      N              YT/YO                        YB/YO                        SF

      1            0.10000E 01                 0,89018E 00                 0.11234E 01
      2            0.10745E 01                 0,81333E 00                 0,13211E 01
      4            0.11653E 01                 0,67895E 00                 0,17164E 01
      6            0.12114E 01                 0,56677E 00                 0.21374E 01
      8            0,12348E 01                 0.47313E 00                 0,26098E 01
     10            0.12466E 01                 0,39496E 00                 0,31564E 01
     12            0.12527E 01                 0.32971E 00                 0.37993E 01
     14            0.12557E 01                 0,27523E 00                 0,45623E 01
     16            0.12573E 01                 0,22976E 00                 0,54721E 01
     18            0.12580E 01                 0.19180E 00                 0.65592E 01
     20            0,12584E 01                 0.16011E 00                 0,78598E 01
     22            0,12586E 01                 0.13366E 00                 0,94170E 01
     24            0.12587E 01                 0.11157E 00                 0.11282E 02
     26            0,12588E 01                 0,93141E-01                 0,13515E 02
     28            0,12588E 01                 0,77752E-01                 0,16190E 02
     30            0.12588E 01                 0.64906E-01                 0.19395E 02
     32            0.12588E 01                 0,54182E-01                 0,23234E 02
     34            0.12589E 01                 0,45230E-01                 0,27832E 02
     36            0.12589E 01                 0.37758E-01                 0.33340E 02
     38            0,12589E 01                 0.31519E-01                 0,39939E 02
     40            0,12589E 01                 0,26312E-01                 0,47844E 02
```

TABLE 11

Experimental Results of Run # 7

$L_1 = 63.35$ $L_2 = 60.90$ Region One

	R^o_{BP} (diluted)	R^o_{TP} (diluted)	$\langle y_{BP} \rangle$ n/yo	$\langle y_{TP} \rangle$ n/yo
1.	-	-	-	-
2.	-	-	-	-
3.	-	-	-	-
4.	+.026	-.240	0.507	1.17
5.	-	-	-	-
6.	-	-	-	-
7.	-	-	-	-
8.	+.077	-.381	0.379	1.53
9.	-	-	-	-
10.	+.093	-.412	0.339	1.61
11.	-	-	-	-
12.	+.115	-.432	0.284	1.66
13.	-	-	-	-
14.	+.125	-.495	0.259	1.82
15.	-	-	-	-
16.	+.138	-.525	0.226	1.89
17.	-	-	-	-
18.	+.140	-	0.221	-
19.	-	-	-	-
20.	+.142	-	0.216	-

```
***************
H=     90.000      B=      0.058
(PHO)UPPER=      0.120      (PHO)LOWER=      0.080
DIST1=    63.347      DIST2=     60.901
(YT/YO)INF=          0.16667E 01      (YB/YO)INF=              0.00000E 00
C1=                0.272            C2=                       0.272
(PHO)LOWER/FEED=      0.400      FEED=      0.200


      N                YT/YO                    YB/YO                    SF

      1             0.10000E 01              0.89018E 00              0.11234E 01
      2             0.10932E 01              0.81333E 00              0.13442E 01
      4             0.12542E 01              0.67895E 00              0.18472E 01
      6             0.13862E 01              0.56677E 00              0.24457E 01
      8             0.14944E 01              0.47313E 00              0.31586E 01
     10             0.15832E 01              0.39496E 00              0.40085E 01
     12             0.16561E 01              0.32971E 00              0.50228E 01
     14             0.17127E 01              0.27523E 00              0.62227E 01
     16             0.17501E 01              0.22976E 00              0.76172E 01
     18             0.17733E 01              0.19180E 00              0.92454E 01
     20             0.17859E 01              0.16011E 00              0.11154E 02
     22             0.17911E 01              0.13366E 00              0.13401E 02
     24             0.17909E 01              0.11157E 00              0.16051E 02
     26             0.17871E 01              0.93141E-01              0.19187E 02
     28             0.17809E 01              0.77752E-01              0.22905E 02
     30             0.17733E 01              0.64906E-01              0.27321E 02
     32             0.17649E 01              0.54182E-01              0.32573E 02
     34             0.17562E 01              0.45230E-01              0.38828E 02
     36             0.17476E 01              0.37758E-01              0.46286E 02
     38             0.17393E 01              0.31519E-01              0.55143E 02
     40             0.17315E 01              0.26312E-01              0.65808E 02
```

TABLE 12

Experimental Results of Run # 8

$L_1 = 60.02$ $L_2 = 59.77$ Region One

	Hot Half Cycle R^o_{BP} (diluted)	R_{TP} (diluted)	$\langle y_{BP} \rangle$ n/yo	$\langle y_{TP} \rangle$ n/yo
1.	-	-	-	-
2.	-	-	-	-
3.	-	-	-	-
4.	-.020	-.208	0.617	1.09
5.	-	-	-	-
6.	+.013	-	0.543	-
7.	-	-	-	-
8.	+.043	-.278	0.476	1.27
9.	-	-	-	-
10.	+.071	-.312	0.414	1.36
11.	-	-	-	-
12.	+.105	-.322	0.338	1.38
13.	-	-	-	-
14.	+.122	-.330	0.300	1.40
15.	-	-	-	-
16.	+.140	-.334	0.260	1.41

```
***************
 H=     90.000      B=      0.058
 (PHO)UPPER=      0.144      (PHO)LOWER=      0.056
 DIST1=    60.022       DIST2=      59.769
 (YT/YO)INF=            0.13889E 01       (YB/YO)INF=            0.00000E 00
 C1=                   0.272             C2=                        0.272
 (PHO)LOWER/FEED=      0.280       FEED=       0.200

      N                YT/YO                     YB/YO                     SF

      1             0.10000E 01               0.89018E 00               0.11234E 01
      2             0.10797E 01               0.81333E 00               0.13275E 01
      4             0.11949E 01               0.67895E 00               0.17599E 01
      6             0.12690E 01               0.56677E 00               0.22390E 01
      8             0.13167E 01               0.47313E 00               0.27829E 01
     10             0.13473E 01               0.39496E 00               0.34113E 01
     12             0.13671E 01               0.32971E 00               0.41463E 01
     14             0.13798E 01               0.27523E 00               0.50131E 01
     16             0.13879E 01               0.22976E 00               0.60408E 01
     18             0.13932E 01               0.19180E 00               0.72638E 01
     20             0.13966E 01               0.16011E 00               0.87225E 01
     22             0.13987E 01               0.13366E 00               0.10465E 02
     24             0.14001E 01               0.11157E 00               0.12549E 02
     26             0.14010E 01               0.93141E-01               0.15042E 02
     28             0.14016E 01               0.77752E-01               0.18027E 02
     30             0.14020E 01               0.64906E-01               0.21600E 02
     32             0.14022E 01               0.54182E-01               0.25880E 02
     34             0.14024E 01               0.45230E-01               0.31005E 02
     36             0.14025E 01               0.37758E-01               0.37145E 02
     38             0.14026E 01               0.31519E-01               0.44498E 02
     40             0.14026E 01               0.26312E-01               0.53307E 02
```

Figure 6

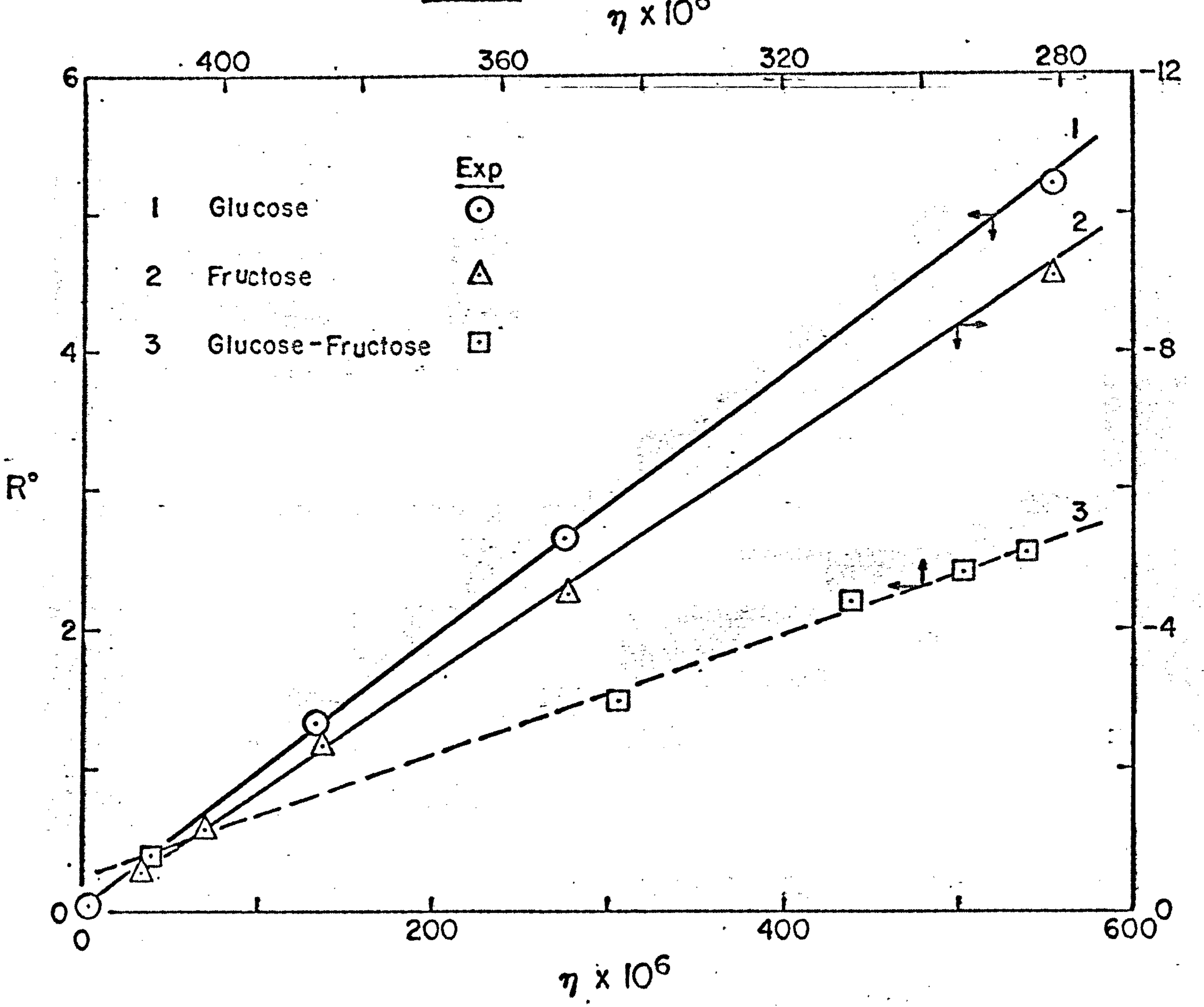
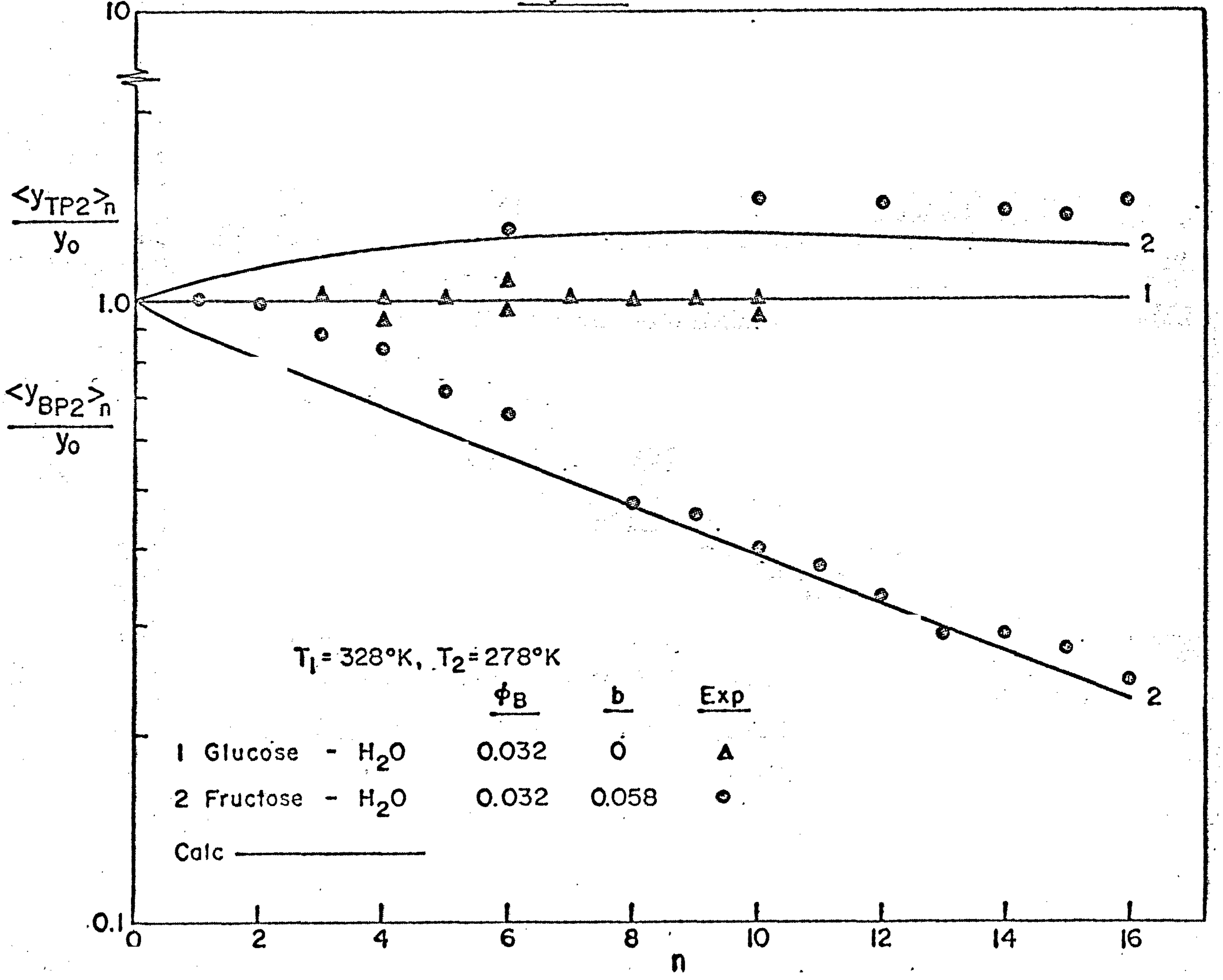
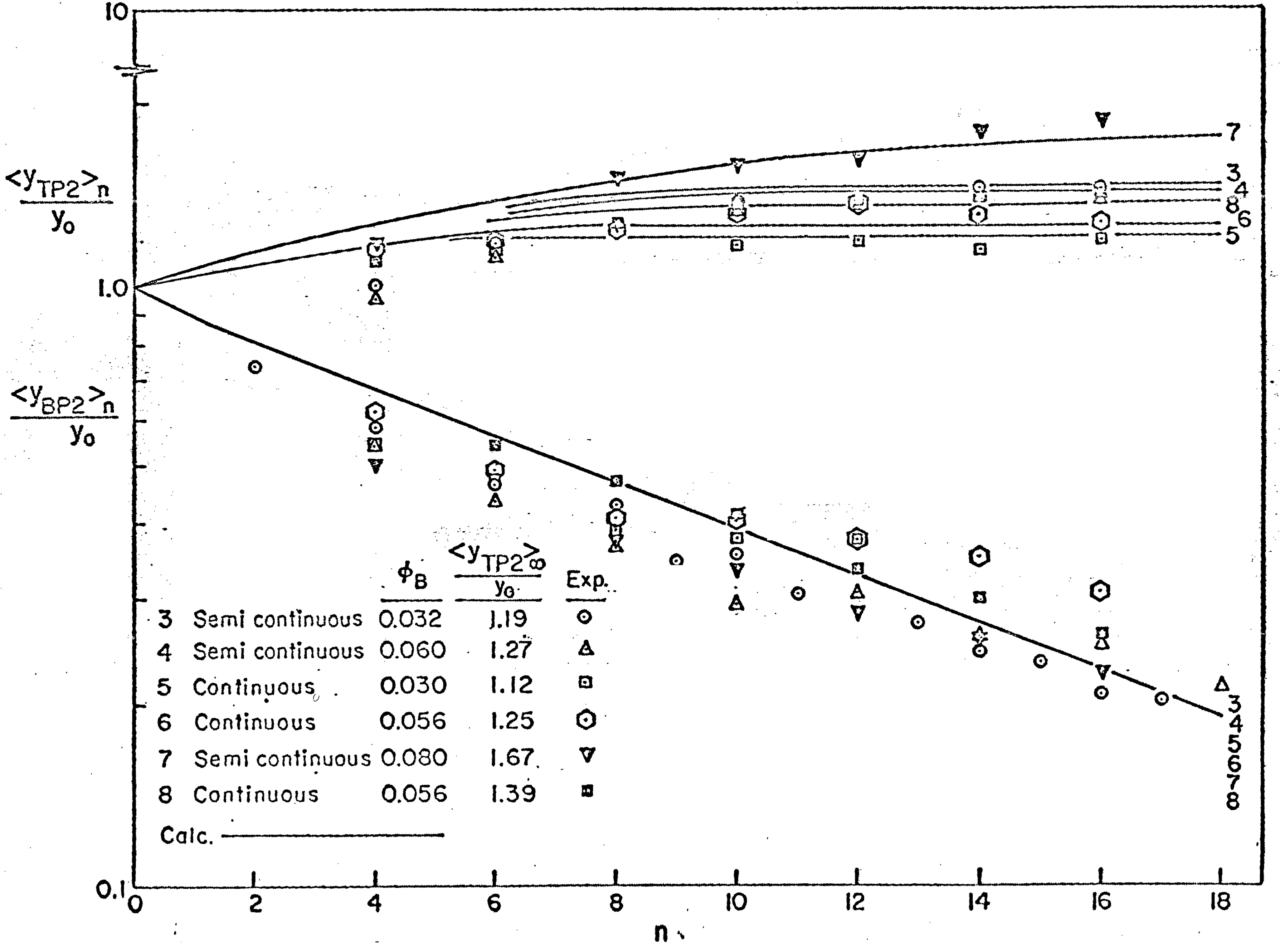

$\eta \times 10^6$
400
360
320
280
6
4
2
0
12
8
4
0
R°
R°
Exp
1 Glucose ⊙
2 Fructose △
3 Glucose-Fructose ⊡
1
2
3
0
200
400
600
$\eta \times 10^6$

Figure 7



10
1.0
0.1
$\frac{\langle y_{TP2} \rangle_n}{y_0}$
$\frac{\langle y_{BP2} \rangle_n}{y_0}$
0
2
4
6
8
10
12
14
16
n
2
1
2
$T_1 = 328°K$, $T_2 = 278°K$
ϕ_B
b
Exp
1 Glucose - H_2O 0.032 0 ▲
2 Fructose - H_2O 0.032 0.058 ⊙
Calc ———

Figure 8



10
1.0
0.1
$\frac{\langle y_{TP2}\rangle_n}{y_o}$
$\frac{\langle y_{BP2}\rangle_n}{y_o}$
0
2
4
6
8
10
12
14
16
18
n
7
3
4
8
6
5
3
4
5
6
7
8
ϕ_B
$\frac{\langle y_{TP2}\rangle_\infty}{y_o}$
Exp.
3 Semi continuous 0.032 1.19 ⊙
4 Semi continuous 0.060 1.27 △
5 Continuous 0.030 1.12 ⊡
6 Continuous 0.056 1.25 ⬡
7 Semi continuous 0.080 1.67 ▽
8 Continuous 0.056 1.39 ▣
Calc. ———

Figure 9

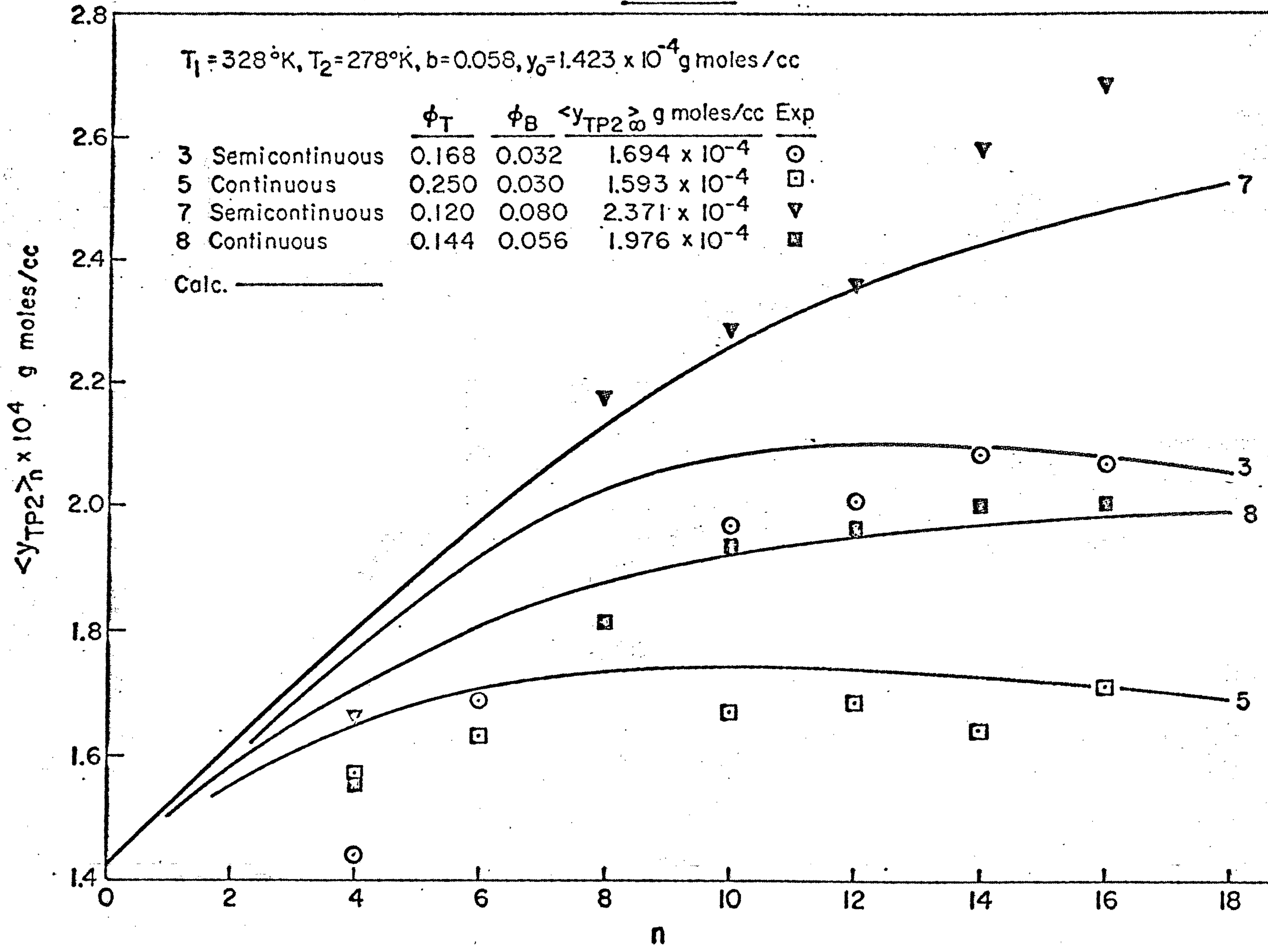

$T_1 = 328°K, T_2 = 278°K, b = 0.058, y_0 = 1.423 \times 10^{-4}$ g moles/cc
ϕ_T ϕ_B $\langle y_{TP2} \rangle_\infty$ g moles/cc Exp
3 Semicontinuous 0.168 0.032 1.694×10^{-4} ⊙
5 Continuous 0.250 0.030 1.593×10^{-4} ⊡
7 Semicontinuous 0.120 0.080 2.371×10^{-4} ▼
8 Continuous 0.144 0.056 1.976×10^{-4} ▣
Calc. ———
$\langle y_{TP2} \rangle_n \times 10^4$ g moles/cc
2.8
2.6
2.4
2.2
2.0
1.8
1.6
1.4
0
2
4
6
8
10
12
14
16
18
n
7
3
8
5

Figure 10

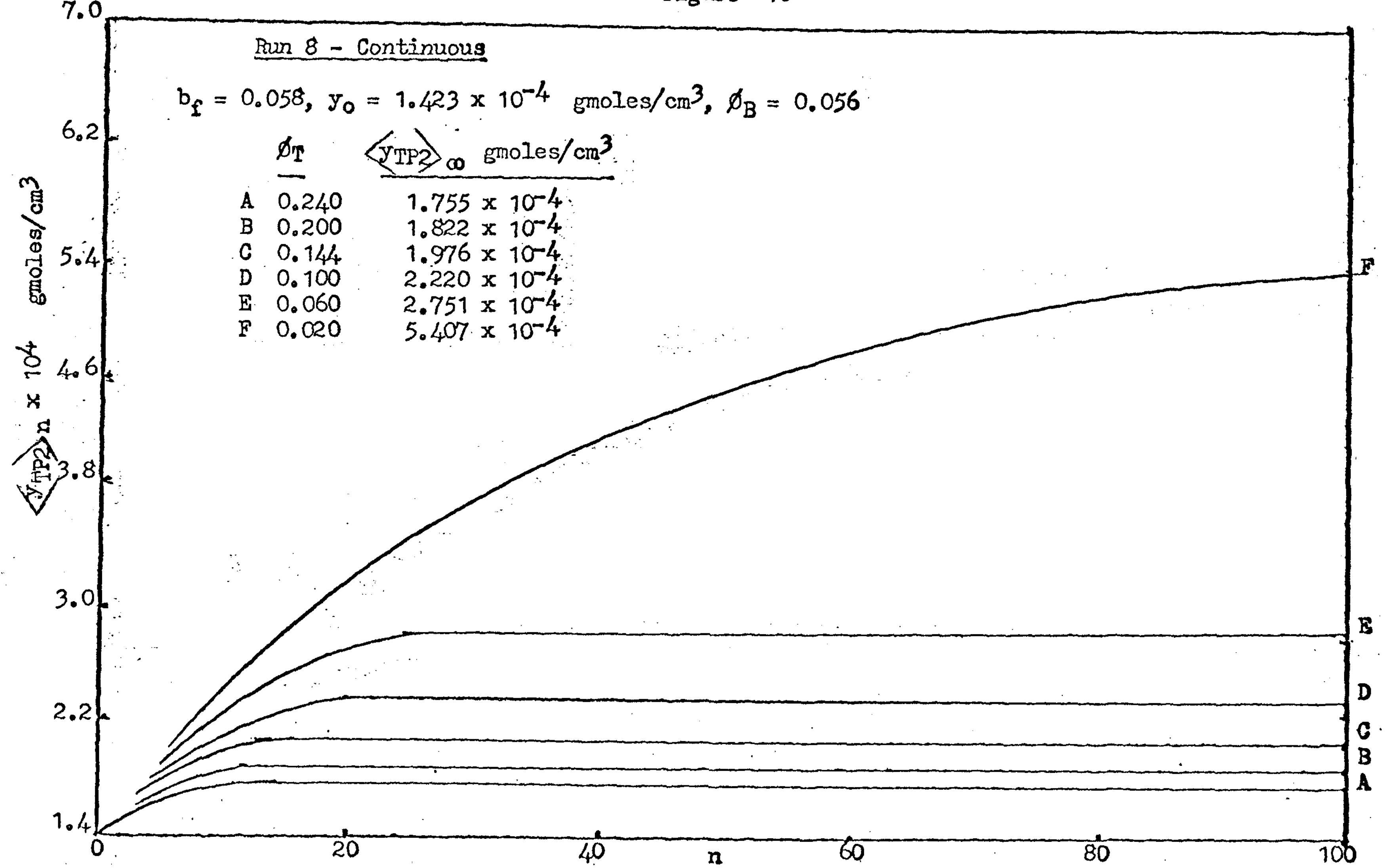
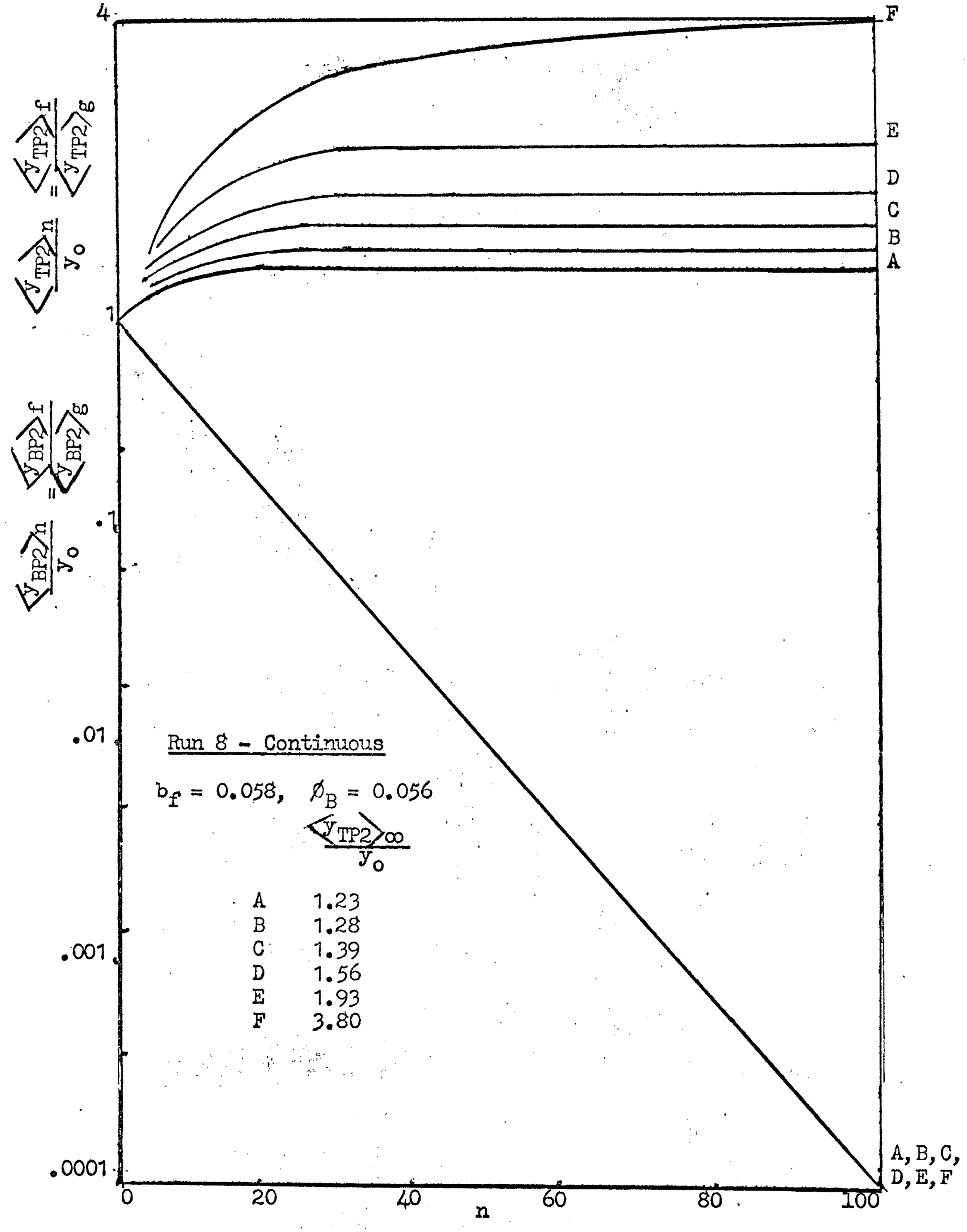

Run 8 - Continuous
$b_f = 0.058$, $y_o = 1.423 \times 10^{-4}$ gmoles/cm^3, $\phi_B = 0.056$
ϕ_T
$\langle y_{TP2} \rangle_\infty$ gmoles/cm^3
A 0.240 1.755×10^{-4}
B 0.200 1.822×10^{-4}
C 0.144 1.976×10^{-4}
D 0.100 2.220×10^{-4}
E 0.060 2.751×10^{-4}
F 0.020 5.407×10^{-4}
$\langle y_{TP2} \rangle_n \times 10^4$ gmoles/cm^3
7.0
6.2
5.4
4.6
3.8
3.0
2.2
1.4
0
20
40
60
80
100
n
F
E
D
C
B
A

Figure 11


4
1
.1
.01
.001
.0001
$\frac{\langle y_{TP2}\rangle_n}{y_o} = \frac{\langle y_{TP2}\rangle_f}{\langle y_{TP2}\rangle_g}$
$\frac{\langle y_{BP2}\rangle_n}{y_o} = \frac{\langle y_{BP2}\rangle_f}{\langle y_{BP2}\rangle_g}$
0
20
40
60
80
100
n
F
E
D
C
B
A
A,B,C,
D,E,F
Run 8 - Continuous
$b_f = 0.058$, $\phi_B = 0.056$
$\frac{\langle y_{TP2}\rangle_\infty}{y_o}$
A 1.23
B 1.28
C 1.39
D 1.56
E 1.93
F 3.80

```
****************
 H=     90.000      B=      0.058
 (PHO)UPPER=      0.240      (PHO)LOWER=      0.056
 DIST1=     60.021      DIST2=      59.769
 (YT/YO)INF=           0.12333E 01      (YB/YO)INF=           0.00000E 00
 C1=                 0.272            C2=                  0.272
 (PHO)LOWER/FEED=      0.189      FEED=      0.296

     N                 YT/YO                          YB/YO                          SF

    1               0.10000E 01                  0.89018E 00                  0.11234E 01
    2               0.10735E 01                  0.81333E 00                  0.13199E 01
    4               0.11603E 01                  0.67895E 00                  0.17089E 01
    6               0.12022E 01                  0.56677E 00                  0.21212E 01
    8               0.12225E 01                  0.47313E 00                  0.25839E 01
   10               0.12324E 01                  0.39496E 00                  0.31202E 01
   12               0.12371E 01                  0.32971E 00                  0.37522E 01
   14               0.12394E 01                  0.27523E 00                  0.45032E 01
   16               0.12406E 01                  0.22976E 00                  0.53994E 01
   18               0.12411E 01                  0.19180E 00                  0.64708E 01
   20               0.12414E 01                  0.16011E 00                  0.77531E 01
   22               0.12415E 01                  0.13366E 00                  0.92886E 01
   24               0.12415E 01                  0.11157E 00                  0.11127E 02
   26               0.12416E 01                  0.93141E-01                  0.13330E 02
   28               0.12416E 01                  0.77752E-01                  0.15969E 02
   30               0.12416E 01                  0.64906E-01                  0.19129E 02
   32               0.12416E 01                  0.54182E-01                  0.22915E 02
   34               0.12416E 01                  0.45230E-01                  0.27451E 02
   36               0.12416E 01                  0.37758E-01                  0.32884E 02
   38               0.12416E 01                  0.31519E-01                  0.39392E 02
   40               0.12416E 01                  0.26312E-01                  0.47188E 02
   42               0.12416E 01                  0.21965E-01                  0.56528E 02
   44               0.12416E 01                  0.18336E-01                  0.67715E 02
   46               0.12416E 01                  0.15306E-01                  0.81117E 02
   48               0.12416E 01                  0.12777E-01                  0.97172E 02
   50               0.12416E 01                  0.10666E-01                  0.11640E 03
   52               0.12416E 01                  0.89040E-02                  0.13944E 03
   54               0.12416E 01                  0.74329E-02                  0.16704E 03
   56               0.12416E 01                  0.62049E-02                  0.20010E 03
   58               0.12416E 01                  0.51797E-02                  0.23971E 03
   60               0.12416E 01                  0.43239E-02                  0.28715E 03
   62               0.12416E 01                  0.36095E-02                  0.34398E 03
   64               0.12416E 01                  0.30132E-02                  0.41206E 03
   66               0.12416E 01                  0.25153E-02                  0.49361E 03
   68               0.12416E 01                  0.20998E-02                  0.59131E 03
   70               0.12416E 01                  0.17528E-02                  0.70834E 03
   72               0.12416E 01                  0.14632E-02                  0.84853E 03
   74               0.12416E 01                  0.12215E-02                  0.10165E 04
   76               0.12416E 01                  0.10197E-02                  0.12176E 04
   78               0.12416E 01                  0.85121E-03                  0.14586E 04
   80               0.12416E 01                  0.71057E-03                  0.17473E 04
   82               0.12416E 01                  0.59317E-03                  0.20932E 04
   84               0.12416E 01                  0.49517E-03                  0.25074E 04
   86               0.12416E 01                  0.41336E-03                  0.30037E 04
   88               0.12416E 01                  0.34506E-03                  0.35982E 04
   90               0.12416E 01                  0.28805E-03                  0.43103E 04
   92               0.12416E 01                  0.24046E-03                  0.51634E 04
   94               0.12416E 01                  0.20073E-03                  0.61854E 04
   96               0.12416E 01                  0.16757E-03                  0.74096E 04
   98               0.12416E 01                  0.13988E-03                  0.88760E 04
  100               0.12416E 01                  0.11677E-03                  0.10633E 05
```

```
***************
 H=    90.000      B=      0.058
 (PHO)UPPER=     0.200     (PHO)LOWER=     0.056
 DIST1=    60.020      DIST2=     59.769
 (YT/YO)INF=           0.12801E 01      (YB/YO)INF=           0.00000E 00
 C1=                 0.272            C2=                 0.272
 (PHO)LOWER/FEED=      0.219     FEED=      0.256

     N                YT/YO                    YB/YO                    SF

    1             0.10000E 01              0.89018E 00              0.11234E 01
    2             0.10760E 01              0.81333E 00              0.13229E 01
    4             0.11734E 01              0.67895E 00              0.17283E 01
    6             0.12265E 01              0.56677E 00              0.21640E 01
    8             0.12554E 01              0.47313E 00              0.26533E 01
   10             0.12711E 01              0.39496E 00              0.32183E 01
   12             0.12797E 01              0.32971E 00              0.38813E 01
   14             0.12844E 01              0.27523E 00              0.46664E 01
   16             0.12869E 01              0.22976E 00              0.56011E 01
   18             0.12883E 01              0.19180E 00              0.67168E 01
   20             0.12890E 01              0.16011E 00              0.80509E 01
   22             0.12895E 01              0.13366E 00              0.96474E 01
   24             0.12897E 01              0.11157E 00              0.11559E 02
   26             0.12898E 01              0.93141E-01              0.13848E 02
   28             0.12899E 01              0.77752E-01              0.16589E 02
   30             0.12899E 01              0.64906E-01              0.19873E 02
   32             0.12899E 01              0.54182E-01              0.23807E 02
   34             0.12899E 01              0.45230E-01              0.28519E 02
   36             0.12899E 01              0.37758E-01              0.34164E 02
   38             0.12899E 01              0.31519E-01              0.40925E 02
   40             0.12899E 01              0.26312E-01              0.49025E 02
   42             0.12899E 01              0.21965E-01              0.58728E 02
   44             0.12899E 01              0.18336E-01              0.70352E 02
   46             0.12899E 01              0.15306E-01              0.84276E 02
   48             0.12899E 01              0.12777E-01              0.10096E 03
   50             0.12899E 01              0.10666E-01              0.12094E 03
   52             0.12899E 01              0.89040E-02              0.14487E 03
   54             0.12899E 01              0.74329E-02              0.17354E 03
   56             0.12899E 01              0.62049E-02              0.20789E 03
   58             0.12899E 01              0.51797E-02              0.24904E 03
   60             0.12899E 01              0.43239E-02              0.29833E 03
   62             0.12899E 01              0.36095E-02              0.35737E 03
   64             0.12899E 01              0.30132E-02              0.42810E 03
   66             0.12899E 01              0.25153E-02              0.51283E 03
   68             0.12899E 01              0.20998E-02              0.61433E 03
   70             0.12899E 01              0.17528E-02              0.73591E 03
   72             0.12899E 01              0.14632E-02              0.88156E 03
   74             0.12899E 01              0.12215E-02              0.10560E 04
   76             0.12899E 01              0.10197E-02              0.12651E 04
   78             0.12899E 01              0.85121E-03              0.15154E 04
   80             0.12899E 01              0.71057E-03              0.18154E 04
   82             0.12899E 01              0.59317E-03              0.21747E 04
   84             0.12899E 01              0.49517E-03              0.26051E 04
   86             0.12899E 01              0.41336E-03              0.31206E 04
   88             0.12899E 01              0.34506E-03              0.37383E 04
   90             0.12899E 01              0.28805E-03              0.44781E 04
   92             0.12899E 01              0.24046E-03              0.53644E 04
   94             0.12899E 01              0.20073E-03              0.64262E 04
   96             0.12899E 01              0.16757E-03              0.76980E 04
   98             0.12899E 01              0.13988E-03              0.92216E 04
  100             0.12899E 01              0.11677E-03              0.11047E 05
```

```
****************
 H=    90.000      B=      0.058
 (PHO)UPPER=      0.144      (PHO)LOWER=      0.056
 DIST1=    60.022      DIST2=     59.769
 (YT/YO)INF=            0.13889E 01      (YB/YO)INF=            0.00000E 00
 C1=                   0.272            C2=                  0.272
 (PHO)LOWER/FEED=      0.280     FEED=      0.200


    N                YT/YO                    YB/YO                  SF

    1             0.10000E 01             0.89018E 00             0.11234E 01
    2             0.10797E 01             0.81333E 00             0.13275E 01
    4             0.11949E 01             0.67895E 00             0.17599E 01
    6             0.12690E 01             0.56677E 00             0.22390E 01
    8             0.13167E 01             0.47313E 00             0.27829E 01
   10             0.13473E 01             0.39496E 00             0.34113E 01
   12             0.13671E 01             0.32971E 00             0.41463E 01
   14             0.13798E 01             0.27523E 00             0.50131E 01
   16             0.13879E 01             0.22976E 00             0.60408E 01
   18             0.13932E 01             0.19180E 00             0.72638E 01
   20             0.13966E 01             0.16011E 00             0.87225E 01
   22             0.13987E 01             0.13366E 00             0.10465E 02
   24             0.14001E 01             0.11157E 00             0.12549E 02
   26             0.14010E 01             0.93141E-01             0.15042E 02
   28             0.14016E 01             0.77752E-01             0.18027E 02
   30             0.14020E 01             0.64906E-01             0.21600E 02
   32             0.14022E 01             0.54182E-01             0.25880E 02
   34             0.14024E 01             0.45230E-01             0.31005E 02
   36             0.14025E 01             0.37758E-01             0.37145E 02
   38             0.14026E 01             0.31519E-01             0.44498E 02
   40             0.14026E 01             0.26312E-01             0.53307E 02
   42             0.14026E 01             0.21965E-01             0.63858E 02
   44             0.14026E 01             0.18336E-01             0.76498E 02
   46             0.14026E 01             0.15306E-01             0.91639E 02
   48             0.14027E 01             0.12777E-01             0.10978E 03
   50             0.14027E 01             0.10666E-01             0.13150E 03
   52             0.14027E 01             0.89040E-02             0.15753E 03
   54             0.14027E 01             0.74329E-02             0.18871E 03
   56             0.14027E 01             0.62049E-02             0.22606E 03
   58             0.14027E 01             0.51797E-02             0.27080E 03
   60             0.14027E 01             0.43239E-02             0.32440E 03
   62             0.14027E 01             0.36095E-02             0.38860E 03
   64             0.14027E 01             0.30132E-02             0.46551E 03
   66             0.14027E 01             0.25153E-02             0.55764E 03
   68             0.14027E 01             0.20998E-02             0.66801E 03
   70             0.14027E 01             0.17528E-02             0.80022E 03
   72             0.14027E 01             0.14632E-02             0.95860E 03
   74             0.14027E 01             0.12215E-02             0.11483E 04
   76             0.14027E 01             0.10197E-02             0.13756E 04
   78             0.14027E 01             0.85121E-03             0.16479E 04
   80             0.14027E 01             0.71057E-03             0.19740E 04
   82             0.14027E 01             0.59317E-03             0.23647E 04
   84             0.14027E 01             0.49517E-03             0.28327E 04
   86             0.14027E 01             0.41336E-03             0.33933E 04
   88             0.14027E 01             0.34506E-03             0.40650E 04
   90             0.14027E 01             0.28805E-03             0.48695E 04
   92             0.14027E 01             0.24046E-03             0.58332E 04
   94             0.14027E 01             0.20073E-03             0.69877E 04
   96             0.14027E 01             0.16757E-03             0.83707E 04
   98             0.14027E 01             0.13988E-03             0.10027E 05
  100             0.14027E 01             0.11677E-03             0.12012E 05
```

```
****************
 H=    90.000      B=      0.058
 (PHO)UPPER=      0.100      (PHO)LOWER=      0.056
 DIST1=    60.021      DIST2=    59.769
 (YT/YO)INF=           0.15601E 01      (YB/YO)INF=           0.00000E 00
 C1=                 0.272           C2=                  0.272
 (PHO)LOWER/FEED=      0.359     FEED=      0.156


    N                 YT/YO                         YB/YO                         SF

    1             0.10000E 01                  0.89018E 00                  0.11234E 01
    2             0.10829E 01                  0.81333E 00                  0.13314E 01
    4             0.12148E 01                  0.67895E 00                  0.17892E 01
    6             0.13117E 01                  0.56677E 00                  0.23143E 01
    8             0.13829E 01                  0.47313E 00                  0.29228E 01
   10             0.14352E 01                  0.39496E 00                  0.36337E 01
   12             0.14736E 01                  0.32971E 00                  0.44693E 01
   14             0.15018E 01                  0.27523E 00                  0.54564E 01
   16             0.15225E 01                  0.22976E 00                  0.66265E 01
   18             0.15377E 01                  0.19180E 00                  0.80174E 01
   20             0.15489E 01                  0.16011E 00                  0.96741E 01
   22             0.15571E 01                  0.13366E 00                  0.11650E 02
   24             0.15632E 01                  0.11157E 00                  0.14010E 02
   26             0.15676E 01                  0.93141E-01                  0.16830E 02
   28             0.15709E 01                  0.77752E-01                  0.20203E 02
   30             0.15732E 01                  0.64906E-01                  0.24239E 02
   32             0.15750E 01                  0.54182E-01                  0.29069E 02
   34             0.15763E 01                  0.45230E-01                  0.34850E 02
   36             0.15772E 01                  0.37758E-01                  0.41773E 02
   38             0.15779E 01                  0.31519E-01                  0.50063E 02
   40             0.15785E 01                  0.26312E-01                  0.59990E 02
   42             0.15788E 01                  0.21965E-01                  0.71881E 02
   44             0.15791E 01                  0.18336E-01                  0.86122E 02
   46             0.15793E 01                  0.15306E-01                  0.10318E 03
   48             0.15795E 01                  0.12777E-01                  0.12361E 03
   50             0.15796E 01                  0.10666E-01                  0.14809E 03
   52             0.15796E 01                  0.89040E-02                  0.17741E 03
   54             0.15797E 01                  0.74329E-02                  0.21253E 03
   56             0.15797E 01                  0.62049E-02                  0.25460E 03
   58             0.15798E 01                  0.51797E-02                  0.30499E 03
   60             0.15798E 01                  0.43239E-02                  0.36536E 03
   62             0.15798E 01                  0.36095E-02                  0.43768E 03
   64             0.15798E 01                  0.30132E-02                  0.52431E 03
   66             0.15798E 01                  0.25153E-02                  0.62808E 03
   68             0.15799E 01                  0.20998E-02                  0.75239E 03
   70             0.15799E 01                  0.17528E-02                  0.90131E 03
   72             0.15799E 01                  0.14632E-02                  0.10797E 04
   74             0.15799E 01                  0.12215E-02                  0.12934E 04
   76             0.15799E 01                  0.10197E-02                  0.15494E 04
   78             0.15799E 01                  0.85121E-03                  0.18560E 04
   80             0.15799E 01                  0.71057E-03                  0.22234E 04
   82             0.15799E 01                  0.59317E-03                  0.26634E 04
   84             0.15799E 01                  0.49517E-03                  0.31906E 04
   86             0.15799E 01                  0.41336E-03                  0.38220E 04
   88             0.15799E 01                  0.34506E-03                  0.45785E 04
   90             0.15799E 01                  0.28805E-03                  0.54846E 04
   92             0.15799E 01                  0.24046E-03                  0.65701E 04
   94             0.15799E 01                  0.20073E-03                  0.78705E 04
   96             0.15799E 01                  0.16757E-03                  0.94282E 04
   98             0.15799E 01                  0.13988E-03                  0.11294E 05
  100             0.15799E 01                  0.11677E-03                  0.13530E 05
```

```
***************
 H=    90.000      B=      0.058
 (PHO)UPPER=      0.060      (PHO)LOWER=      0.056
 DIST1=    60.021      DIST2=     59.769
 (YT/YO)INF=            0.19335E 01      (YB/YO)INF=           0.00000E 00
 C1=                   0.272        C2=                  0.272
 (PHO)LOWER/FEED=      0.483      FEED=      0.116

     N              YT/YO                     YB/YO                    SF

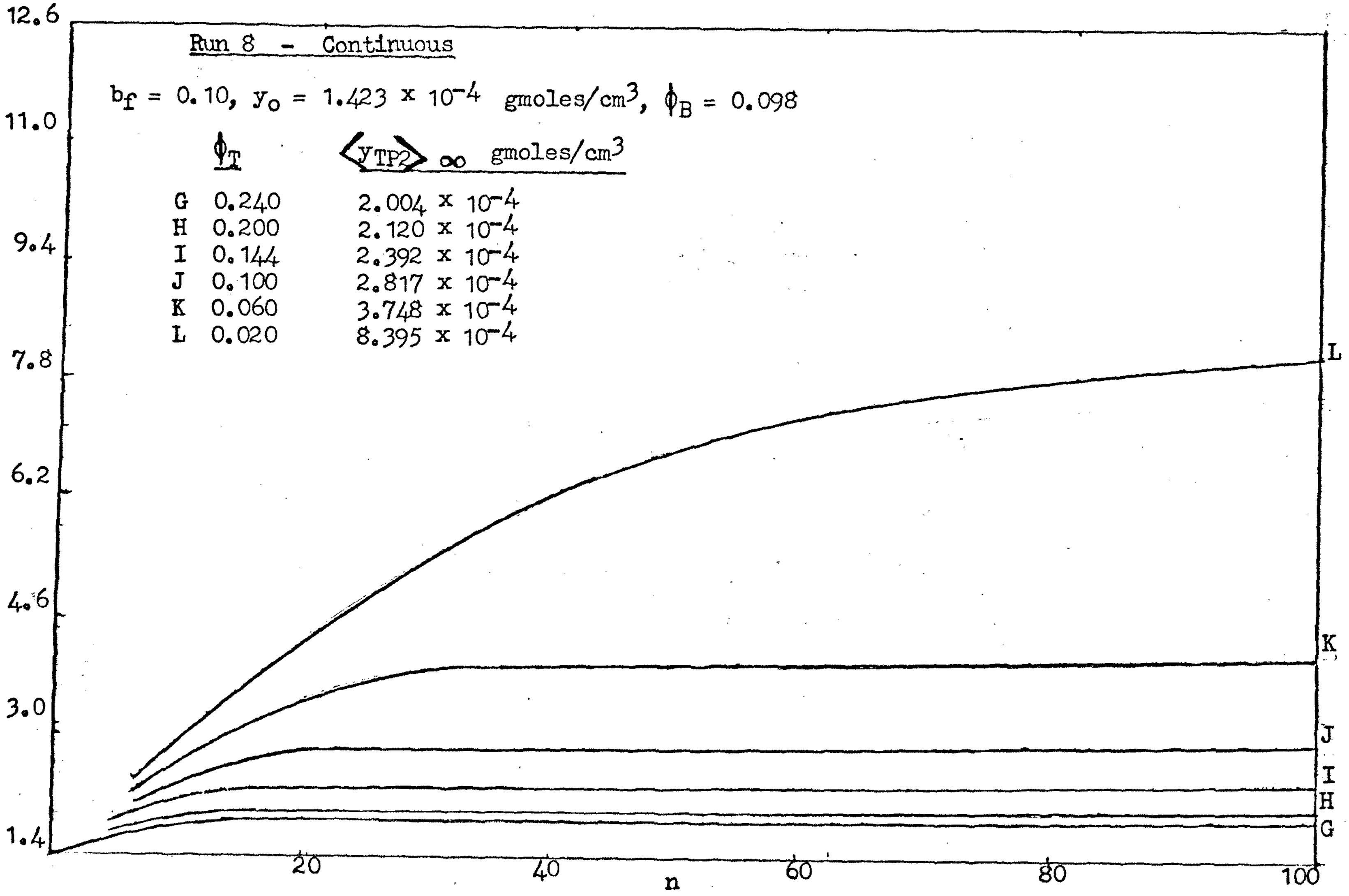
    1            0.10000E 01              0.89018E 00             0.11234E 01
    2            0.10860E 01              0.81333E 00             0.13353E 01
    4            0.12357E 01              0.67895E 00             0.18201E 01
    6            0.13600E 01              0.56677E 00             0.23996E 01
    8            0.14632E 01              0.47313E 00             0.30925E 01
   10            0.15488E 01              0.39496E 00             0.39213E 01
   12            0.16198E 01              0.32971E 00             0.49128E 01
   14            0.16788E 01              0.27523E 00             0.60994E 01
   16            0.17277E 01              0.22976E 00             0.75196E 01
   18            0.17683E 01              0.19180E 00             0.92195E 01
   20            0.18020E 01              0.16011E 00             0.11255E 02
   22            0.18300E 01              0.13366E 00             0.13692E 02
   24            0.18532E 01              0.11157E 00             0.16609E 02
   26            0.18725E 01              0.93141E-01             0.20104E 02
   28            0.18885E 01              0.77752E-01             0.24288E 02
   30            0.19017E 01              0.64906E-01             0.29300E 02
   32            0.19127E 01              0.54182E-01             0.35302E 02
   34            0.19219E 01              0.45230E-01             0.42491E 02
   36            0.19295E 01              0.37758E-01             0.51101E 02
   38            0.19358E 01              0.31519E-01             0.61415E 02
   40            0.19410E 01              0.26312E-01             0.73769E 02
   42            0.19453E 01              0.21965E-01             0.88567E 02
   44            0.19489E 01              0.18336E-01             0.10629E 03
   46            0.19519E 01              0.15306E-01             0.12752E 03
   48            0.19544E 01              0.12777E-01             0.15296E 03
   50            0.19565E 01              0.10666E-01             0.18342E 03
   52            0.19582E 01              0.89040E-02             0.21992E 03
   54            0.19596E 01              0.74329E-02             0.26363E 03
   56            0.19608E 01              0.62049E-02             0.31600E 03
   58            0.19617E 01              0.51797E-02             0.37873E 03
   60            0.19625E 01              0.43239E-02             0.45388E 03
   62            0.19632E 01              0.36095E-02             0.54389E 03
   64            0.19638E 01              0.30132E-02             0.65173E 03
   66            0.19642E 01              0.25153E-02             0.78090E 03
   68            0.19646E 01              0.20998E-02             0.93564E 03
   70            0.19649E 01              0.17528E-02             0.11210E 04
   72            0.19652E 01              0.14632E-02             0.13430E 04
   74            0.19654E 01              0.12215E-02             0.16090E 04
   76            0.19656E 01              0.10197E-02             0.19277E 04
   78            0.19658E 01              0.85121E-03             0.23094E 04
   80            0.19659E 01              0.71057E-03             0.27666E 04
   82            0.19660E 01              0.59317E-03             0.33144E 04
   84            0.19661E 01              0.49517E-03             0.39705E 04
   86            0.19661E 01              0.41336E-03             0.47565E 04
   88            0.19662E 01              0.34506E-03             0.56981E 04
   90            0.19662E 01              0.28805E-03             0.68260E 04
   92            0.19663E 01              0.24046E-03             0.81772E 04
   94            0.19663E 01              0.20073E-03             0.97957E 04
   96            0.19664E 01              0.16757E-03             0.11735E 05
   98            0.19664E 01              0.13988E-03             0.14057E 05
  100            0.19664E 01              0.11677E-03             0.16840E 05
```

```
***************
 H=     90.000      B=      0.058
 (PHO)UPPER=      0.020      (PHO)LOWER=      0.056
 DIST1=    60.022      DIST2=     59.769
 (YT/YO)INF=          0.37994E 01      (YB/YO)INF=          0.00000E 00
 C1=                  0.272          C2=                  0.272
 (PHO)LOWER/FEED=      0.737      FEED=      0.076


     N              YT/YO                     YB/YO                     SF

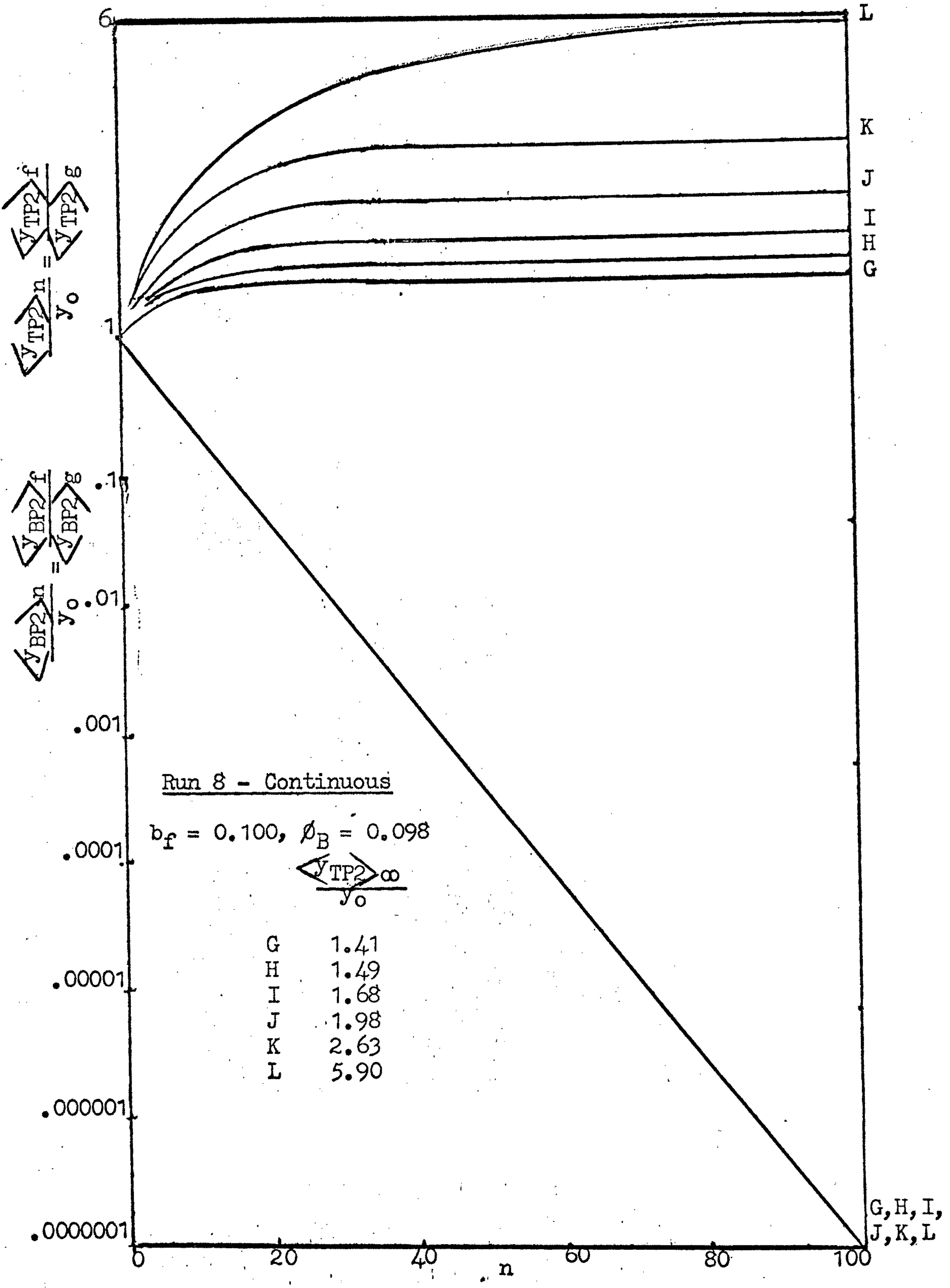
     1            0.10000E 01              0.89018E 00              0.11234E 01
     2            0.10894E 01              0.81333E 00              0.13394E 01
     4            0.12600E 01              0.67895E 00              0.18558E 01
     6            0.14202E 01              0.56677E 00              0.25057E 01
     8            0.15707E 01              0.47313E 00              0.33197E 01
    10            0.17120E 01              0.39496E 00              0.43347E 01
    12            0.18448E 01              0.32971E 00              0.55953E 01
    14            0.19695E 01              0.27523E 00              0.71558E 01
    16            0.20867E 01              0.22976E 00              0.90819E 01
    18            0.21967E 01              0.19180E 00              0.11453E 02
    20            0.23000E 01              0.16011E 00              0.14365E 02
    22            0.23971E 01              0.13366E 00              0.17935E 02
    24            0.24883E 01              0.11157E 00              0.22302E 02
    26            0.25739E 01              0.93141E-01              0.27635E 02
    28            0.26544E 01              0.77752E-01              0.34139E 02
    30            0.27299E 01              0.64906E-01              0.42060E 02
    32            0.28009E 01              0.54182E-01              0.51694E 02
    34            0.28676E 01              0.45230E-01              0.63399E 02
    36            0.29302E 01              0.37758E-01              0.77605E 02
    38            0.29890E 01              0.31519E-01              0.94831E 02
    40            0.30442E 01              0.26312E-01              0.11570E 03
    42            0.30961E 01              0.21965E-01              0.14096E 03
    44            0.31449E 01              0.18336E-01              0.17152E 03
    46            0.31906E 01              0.15306E-01              0.20845E 03
    48            0.32336E 01              0.12777E-01              0.25308E 03
    50            0.32740E 01              0.10666E-01              0.30695E 03
    52            0.33120E 01              0.89040E-02              0.37196E 03
    54            0.33476E 01              0.74329E-02              0.45037E 03
    56            0.33811E 01              0.62049E-02              0.54490E 03
    58            0.34125E 01              0.51797E-02              0.65882E 03
    60            0.34420E 01              0.43239E-02              0.79604E 03
    62            0.34698E 01              0.36095E-02              0.96128E 03
    64            0.34958E 01              0.30132E-02              0.11602E 04
    66            0.35203E 01              0.25153E-02              0.13995E 04
    68            0.35433E 01              0.20998E-02              0.16875E 04
    70            0.35649E 01              0.17528E-02              0.20338E 04
    72            0.35851E 01              0.14632E-02              0.24501E 04
    74            0.36042E 01              0.12215E-02              0.29506E 04
    76            0.36221E 01              0.10197E-02              0.35522E 04
    78            0.36389E 01              0.85121E-03              0.42749E 04
    80            0.36547E 01              0.71057E-03              0.51433E 04
    82            0.36695E 01              0.59317E-03              0.61862E 04
    84            0.36834E 01              0.49517E-03              0.74387E 04
    86            0.36965E 01              0.41336E-03              0.89426E 04
    88            0.37088E 01              0.34506E-03              0.10748E 05
    90            0.37203E 01              0.28805E-03              0.12915E 05
    92            0.37311E 01              0.24046E-03              0.15517E 05
    94            0.37413E 01              0.20073E-03              0.18638E 05
    96            0.37509E 01              0.16757E-03              0.22384E 05
    98            0.37599E 01              0.13988E-03              0.26879E 05
   100            0.37683E 01              0.11677E-03              0.32271E 05
```

Figure 12



Run 8 - Continuous
b_f = 0.10, y_o = 1.423 x 10^{-4} gmoles/cm^3, ϕ_B = 0.098
ϕ_T
$\langle y_{TP2} \rangle_\infty$ gmoles/cm^3
G 0.240 2.004 x 10^{-4}
H 0.200 2.120 x 10^{-4}
I 0.144 2.392 x 10^{-4}
J 0.100 2.817 x 10^{-4}
K 0.060 3.748 x 10^{-4}
L 0.020 8.395 x 10^{-4}
12.6
11.0
9.4
7.8
6.2
4.6
3.0
1.4
20
40
60
80
100
n
L
K
J
I
H
G

Figure 13


6
1
.1
.01
.001
.0001
.00001
.000001
.0000001
$\frac{\langle y_{TP2}\rangle_n}{y_o} = \frac{\langle y_{TP2}\rangle_f}{\langle y_{TP2}\rangle_g}$
$\frac{\langle y_{BP2}\rangle_n}{y_o} = \frac{\langle y_{BP2}\rangle_f}{\langle y_{BP2}\rangle_g}$
0
20
40
60
80
100
n
L
K
J
I
H
G
G,H,I,
J,K,L
Run 8 - Continuous
$b_f = 0.100$, $\phi_B = 0.098$
$\frac{\langle y_{TP2}\rangle_\infty}{y_o}$
G 1.41
H 1.49
I 1.68
J 1.98
K 2.63
L 5.90

```
***************
 H=     90.000      B=      0.100
 (PHO)UPPER=      0.240      (PHO)LOWER=      0.098
 DIST1=     60.021      DIST2=      59.778
 (YT/YO)INF=             0.14083E 01      (YB/YO)INF=             0.00000E 00
 C1=                   0.272             C2=                    0.272
 (PHO)LOWER/FEED=      0.290      FEED=      0.338

     N               YT/YO                      YB/YO                      SF

     1            0.10000E 01                0.81818E 00              0.12222E 01
     2            0.11266E 01                0.70123E 00              0.16065E 01
     4            0.12759E 01                0.51509E 00              0.24770E 01
     6            0.13481E 01                0.37836E 00              0.35630E 01
     8            0.13831E 01                0.27793E 00              0.49764E 01
    10            0.14000E 01                0.20415E 00              0.68576E 01
    12            0.14082E 01                0.14996E 00              0.93904E 01
    14            0.14122E 01                0.11015E 00              0.12820E 02
    16            0.14141E 01                0.80915E-01              0.17476E 02
    18            0.14150E 01                0.59436E-01              0.23807E 02
    20            0.14155E 01                0.43659E-01              0.32421E 02
    22            0.14157E 01                0.32070E-01              0.44143E 02
    24            0.14158E 01                0.23557E-01              0.60100E 02
    26            0.14158E 01                0.17304E-01              0.81821E 02
    28            0.14159E 01                0.12711E-01              0.11139E 03
    30            0.14159E 01                0.93367E-02              0.15165E 03
    32            0.14159E 01                0.68583E-02              0.20645E 03
    34            0.14159E 01                0.50378E-02              0.28105E 03
    36            0.14159E 01                0.37005E-02              0.38261E 03
    38            0.14159E 01                0.27182E-02              0.52083E 03
    40            0.14159E 01                0.19967E-02              0.70911E 03
    42            0.14159E 01                0.14667E-02              0.96536E 03
    44            0.14159E 01                0.10774E-02              0.13142E 04
    46            0.14159E 01                0.79137E-03              0.17891E 04
    48            0.14159E 01                0.58131E-03              0.24357E 04
    50            0.14159E 01                0.42700E-03              0.33159E 04
    52            0.14159E 01                0.31365E-03              0.45141E 04
    54            0.14159E 01                0.23040E-03              0.61454E 04
    56            0.14159E 01                0.16924E-03              0.83662E 04
    58            0.14159E 01                0.12431E-03              0.11389E 05
    60            0.14159E 01                0.91316E-04              0.15505E 05
    62            0.14159E 01                0.67076E-04              0.21108E 05
    64            0.14159E 01                0.49271E-04              0.28736E 05
    66            0.14159E 01                0.36192E-04              0.39121E 05
    68            0.14159E 01                0.26585E-04              0.53258E 05
    70            0.14159E 01                0.19528E-04              0.72504E 05
    72            0.14159E 01                0.14345E-04              0.98705E 05
    74            0.14159E 01                0.10537E-04              0.13437E 06
    76            0.14159E 01                0.77399E-05              0.18293E 06
    78            0.14159E 01                0.56854E-05              0.24904E 06
    80            0.14159E 01                0.41762E-05              0.33903E 06
    82            0.14159E 01                0.30676E-05              0.46155E 06
    84            0.14159E 01                0.22534E-05              0.62834E 06
    86            0.14159E 01                0.16552E-05              0.85541E 06
    88            0.14159E 01                0.12158E-05              0.11645E 07
    90            0.14159E 01                0.89310E-06              0.15854E 07
    92            0.14159E 01                0.65603E-06              0.21582E 07
    94            0.14159E 01                0.48189E-06              0.29382E 07
    96            0.14159E 01                0.35397E-06              0.39999E 07
    98            0.14159E 01                0.26001E-06              0.54454E 07
   100            0.14159E 01                0.19099E-06              0.74132E 07
```

```
****************
 H=    90.000      B=      0.100
 (PHO)UPPER=      0.200      (PHO)LOWER=      0.098
 DIST1=    60.018      DIST2=     59.778
 (YT/YO)INF=          0.14901E 01      (YB/YO)INF=          0.00000E 00
 C1=                0.272            C2=                    0.272
 (PHO)LOWER/FEED=     0.329      FEED=     0.298


    N                 YT/YO                      YB/YO                      SF

    1              0.10000E 01              0.81818E 00              0.12222E 01
    2              0.11308E 01              0.70123E 00              0.16126E 01
    4              0.12985E 01              0.51509E 00              0.25210E 01
    6              0.13899E 01              0.37836E 00              0.36734E 01
    8              0.14396E 01              0.27793E 00              0.51799E 01
   10              0.14667E 01              0.20415E 00              0.71844E 01
   12              0.14815E 01              0.14996E 00              0.98791E 01
   14              0.14895E 01              0.11015E 00              0.13522E 02
   16              0.14939E 01              0.80915E-01              0.18463E 02
   18              0.14963E 01              0.59436E-01              0.25174E 02
   20              0.14976E 01              0.43659E-01              0.34301E 02
   22              0.14983E 01              0.32070E-01              0.46719E 02
   24              0.14987E 01              0.23557E-01              0.63618E 02
   26              0.14989E 01              0.17304E-01              0.86620E 02
   28              0.14990E 01              0.12711E-01              0.11793E 03
   30              0.14990E 01              0.93367E-02              0.16055E 03
   32              0.14991E 01              0.68583E-02              0.21858E 03
   34              0.14991E 01              0.50378E-02              0.29757E 03
   36              0.14991E 01              0.37005E-02              0.40511E 03
   38              0.14991E 01              0.27182E-02              0.55150E 03
   40              0.14991E 01              0.19967E-02              0.75080E 03
   42              0.14991E 01              0.14667E-02              0.10221E 04
   44              0.14991E 01              0.10774E-02              0.13915E 04
   46              0.14991E 01              0.79137E-03              0.18943E 04
   48              0.14991E 01              0.58131E-03              0.25789E 04
   50              0.14991E 01              0.42700E-03              0.35108E 04
   52              0.14991E 01              0.31365E-03              0.47795E 04
   54              0.14991E 01              0.23040E-03              0.65067E 04
   56              0.14991E 01              0.16924E-03              0.88581E 04
   58              0.14991E 01              0.12431E-03              0.12059E 05
   60              0.14991E 01              0.91316E-04              0.16417E 05
   62              0.14991E 01              0.67076E-04              0.22349E 05
   64              0.14991E 01              0.49271E-04              0.30426E 05
   66              0.14991E 01              0.36192E-04              0.41421E 05
   68              0.14991E 01              0.26585E-04              0.56389E 05
   70              0.14991E 01              0.19528E-04              0.76767E 05
   72              0.14991E 01              0.14345E-04              0.10451E 06
   74              0.14991E 01              0.10537E-04              0.14227E 06
   76              0.14991E 01              0.77399E-05              0.19369E 06
   78              0.14991E 01              0.56854E-05              0.26368E 06
   80              0.14991E 01              0.41762E-05              0.35897E 06
   82              0.14991E 01              0.30676E-05              0.48869E 06
   84              0.14991E 01              0.22534E-05              0.66529E 06
   86              0.14991E 01              0.16552E-05              0.90570E 06
   88              0.14991E 01              0.12158E-05              0.12330E 07
   90              0.14991E 01              0.89310E-06              0.16786E 07
   92              0.14991E 01              0.65603E-06              0.22851E 07
   94              0.14991E 01              0.48189E-06              0.31109E 07
   96              0.14991E 01              0.35397E-06              0.42351E 07
   98              0.14991E 01              0.26001E-06              0.57656E 07
  100              0.14991E 01              0.19099E-06              0.78491E 07
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***************
 H=    90.000      B=      0.100
 (PHO)UPPER=      0.144      (PHO)LOWER=      0.098
 DIST1=    60.018      DIST2=     59.778
 (YT/YO)INF=             0.16807E 01      (YB/YO)INF=             0.00000E 00
 C1=                    0.272             C2=                    0.272
 (PHO)LOWER/FEED=      0.405      FEED=      0.242


     N                  YT/YO                        YB/YO                        SF

    1              0.10000E 01                 0.81818E 00                 0.12222E 01
    2              0.11372E 01                 0.70123E 00                 0.16217E 01
    4              0.13355E 01                 0.51509E 00                 0.25927E 01
    6              0.14631E 01                 0.37836E 00                 0.38669E 01
    8              0.15452E 01                 0.27793E 00                 0.55596E 01
   10              0.15980E 01                 0.20415E 00                 0.78273E 01
   12              0.16319E 01                 0.14996E 00                 0.10882E 02
   14              0.16538E 01                 0.11015E 00                 0.15013E 02
   16              0.16679E 01                 0.80915E-01                 0.20613E 02
   18              0.16769E 01                 0.59436E-01                 0.28214E 02
   20              0.16827E 01                 0.43659E-01                 0.38543E 02
   22              0.16865E 01                 0.32070E-01                 0.52587E 02
   24              0.16889E 01                 0.23557E-01                 0.71693E 02
   26              0.16904E 01                 0.17304E-01                 0.97691E 02
   28              0.16914E 01                 0.12711E-01                 0.13307E 03
   30              0.16921E 01                 0.93367E-02                 0.18123E 03
   32              0.16925E 01                 0.68583E-02                 0.24678E 03
   34              0.16928E 01                 0.50378E-02                 0.33601E 03
   36              0.16929E 01                 0.37005E-02                 0.45748E 03
   38              0.16930E 01                 0.27182E-02                 0.62284E 03
   40              0.16931E 01                 0.19967E-02                 0.84796E 03
   42              0.16931E 01                 0.14667E-02                 0.11544E 04
   44              0.16932E 01                 0.10774E-02                 0.15716E 04
   46              0.16932E 01                 0.79137E-03                 0.21396E 04
   48              0.16932E 01                 0.58131E-03                 0.29128E 04
   50              0.16932E 01                 0.42700E-03                 0.39654E 04
   52              0.16932E 01                 0.31365E-03                 0.53984E 04
   54              0.16932E 01                 0.23040E-03                 0.73492E 04
   56              0.16932E 01                 0.16924E-03                 0.10005E 05
   58              0.16932E 01                 0.12431E-03                 0.13620E 05
   60              0.16932E 01                 0.91316E-04                 0.18543E 05
   62              0.16932E 01                 0.67076E-04                 0.25243E 05
   64              0.16932E 01                 0.49271E-04                 0.34365E 05
   66              0.16932E 01                 0.36192E-04                 0.46784E 05
   68              0.16932E 01                 0.26585E-04                 0.63691E 05
   70              0.16932E 01                 0.19528E-04                 0.86707E 05
   72              0.16932E 01                 0.14345E-04                 0.11804E 06
   74              0.16932E 01                 0.10537E-04                 0.16070E 06
   76              0.16932E 01                 0.77399E-05                 0.21877E 06
   78              0.16932E 01                 0.56854E-05                 0.29782E 06
   80              0.16932E 01                 0.41762E-05                 0.40545E 06
   82              0.16932E 01                 0.30676E-05                 0.55196E 06
   84              0.16932E 01                 0.22534E-05                 0.75143E 06
   86              0.16932E 01                 0.16552E-05                 0.10230E 07
   88              0.16932E 01                 0.12158E-05                 0.13926E 07
   90              0.16932E 01                 0.89310E-06                 0.18959E 07
   92              0.16932E 01                 0.65603E-06                 0.25810E 07
   94              0.16932E 01                 0.48189E-06                 0.35137E 07
   96              0.16932E 01                 0.35397E-06                 0.47835E 07
   98              0.16932E 01                 0.26001E-06                 0.65121E 07
  100              0.16932E 01                 0.19099E-06                 0.88654E 07
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***************
 H=     90.000      B=      0.100
 (PHO)UPPER=      0.100      (PHO)LOWER=      0.098
 DIST1=    60.021      DIST2=    59.778
 (YT/YO)INF=          0.19798E 01      (YB/YO)INF=          0.00000E 00
 C1=                0.272           C2=                0.272
 (PHO)LOWER/FEED=      0.495      FEED=      0.198

     N               YT/YO                   YB/YO                   SF

     1            0.10000E 01            0.81818E 00            0.12222E 01
     2            0.11427E 01            0.70123E 00            0.16295E 01
     4            0.13698E 01            0.51509E 00            0.26592E 01
     6            0.15366E 01            0.37836E 00            0.40610E 01
     8            0.16591E 01            0.27793E 00            0.59694E 01
    10            0.17491E 01            0.20415E 00            0.85674E 01
    12            0.18152E 01            0.14996E 00            0.12104E 02
    14            0.18637E 01            0.11015E 00            0.16919E 02
    16            0.18994E 01            0.80915E-01            0.23474E 02
    18            0.19256E 01            0.59436E-01            0.32398E 02
    20            0.19448E 01            0.43659E-01            0.44546E 02
    22            0.19590E 01            0.32070E-01            0.61084E 02
    24            0.19693E 01            0.23557E-01            0.83599E 02
    26            0.19770E 01            0.17304E-01            0.11425E 03
    28            0.19826E 01            0.12711E-01            0.15598E 03
    30            0.19867E 01            0.93367E-02            0.21278E 03
    32            0.19897E 01            0.68583E-02            0.29012E 03
    34            0.19919E 01            0.50378E-02            0.39540E 03
    36            0.19936E 01            0.37005E-02            0.53872E 03
    38            0.19948E 01            0.27182E-02            0.73384E 03
    40            0.19956E 01            0.19967E-02            0.99947E 03
    42            0.19963E 01            0.14667E-02            0.13611E 04
    44            0.19968E 01            0.10774E-02            0.18534E 04
    46            0.19971E 01            0.79137E-03            0.25236E 04
    48            0.19974E 01            0.58131E-03            0.34360E 04
    50            0.19976E 01            0.42700E-03            0.46781E 04
    52            0.19977E 01            0.31365E-03            0.63691E 04
    54            0.19978E 01            0.23040E-03            0.86711E 04
    56            0.19979E 01            0.16924E-03            0.11805E 05
    58            0.19979E 01            0.12431E-03            0.16071E 05
    60            0.19980E 01            0.91316E-04            0.21880E 05
    62            0.19980E 01            0.67076E-04            0.29787E 05
    64            0.19980E 01            0.49271E-04            0.40551E 05
    66            0.19980E 01            0.36192E-04            0.55206E 05
    68            0.19980E 01            0.26585E-04            0.75156E 05
    70            0.19980E 01            0.19528E-04            0.10232E 06
    72            0.19981E 01            0.14345E-04            0.13929E 06
    74            0.19981E 01            0.10537E-04            0.18963E 06
    76            0.19981E 01            0.77399E-05            0.25815E 06
    78            0.19981E 01            0.56854E-05            0.35144E 06
    80            0.19981E 01            0.41762E-05            0.47844E 06
    82            0.19981E 01            0.30676E-05            0.65134E 06
    84            0.19981E 01            0.22534E-05            0.88671E 06
    86            0.19981E 01            0.16552E-05            0.12071E 07
    88            0.19981E 01            0.12158E-05            0.16434E 07
    90            0.19981E 01            0.89310E-06            0.22372E 07
    92            0.19981E 01            0.65603E-06            0.30457E 07
    94            0.19981E 01            0.48189E-06            0.41463E 07
    96            0.19981E 01            0.35397E-06            0.56447E 07
    98            0.19981E 01            0.26001E-06            0.76845E 07
   100            0.19981E 01            0.19099E-06            0.10461E 08
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***************
 H=    90.000      B=      0.100
 (PHO)UPPER=      0.060      (PHO)LOWER=      0.098
 DIST1=    60.019      DIST2=    59.778
 (YT/YO)INF=            0.26337E 01      (YB/YO)INF=          0.00000E 00
 C1=                  0.272            C2=                  0.272
 (PHO)LOWER/FEED=      0.620      FEED=      0.158

    N              YT/YO                    YB/YO                    SF

   1             0.10000E 01              0.81818E 00              0.12222E 01
   2             0.11481E 01              0.70123E 00              0.16372E 01
   4             0.14058E 01              0.51509E 00              0.27293E 01
   6             0.16198E 01              0.37836E 00              0.42810E 01
   8             0.17973E 01              0.27793E 00              0.64669E 01
  10             0.19447E 01              0.20415E 00              0.95256E 01
  12             0.20670E 01              0.14996E 00              0.13783E 02
  14             0.21685E 01              0.11015E 00              0.19686E 02
  16             0.22527E 01              0.80915E-01              0.27841E 02
  18             0.23226E 01              0.59436E-01              0.39078E 02
  20             0.23807E 01              0.43659E-01              0.54529E 02
  22             0.24288E 01              0.32070E-01              0.75735E 02
  24             0.24688E 01              0.23557E-01              0.10480E 03
  26             0.25020E 01              0.17304E-01              0.14459E 03
  28             0.25295E 01              0.12711E-01              0.19901E 03
  30             0.25523E 01              0.93367E-02              0.27337E 03
  32             0.25713E 01              0.68583E-02              0.37492E 03
  34             0.25870E 01              0.50378E-02              0.51353E 03
  36             0.26001E 01              0.37005E-02              0.70263E 03
  38             0.26110E 01              0.27182E-02              0.96053E 03
  40             0.26199E 01              0.19967E-02              0.13121E 04
  42             0.26274E 01              0.14667E-02              0.17914E 04
  44             0.26336E 01              0.10774E-02              0.24445E 04
  46             0.26388E 01              0.79137E-03              0.33344E 04
  48             0.26430E 01              0.58131E-03              0.45467E 04
  50             0.26466E 01              0.42700E-03              0.61980E 04
  52             0.26495E 01              0.31365E-03              0.84472E 04
  54             0.26519E 01              0.23040E-03              0.11510E 05
  56             0.26540E 01              0.16924E-03              0.15682E 05
  58             0.26557E 01              0.12431E-03              0.21362E 05
  60             0.26570E 01              0.91316E-04              0.29097E 05
  62             0.26582E 01              0.67076E-04              0.39630E 05
  64             0.26592E 01              0.49271E-04              0.53970E 05
  66             0.26600E 01              0.36192E-04              0.73495E 05
  68             0.26606E 01              0.26585E-04              0.10008E 06
  70             0.26612E 01              0.19528E-04              0.13627E 06
  72             0.26616E 01              0.14345E-04              0.18555E 06
  74             0.26620E 01              0.10537E-04              0.25264E 06
  76             0.26623E 01              0.77399E-05              0.34397E 06
  78             0.26626E 01              0.56854E-05              0.46832E 06
  80             0.26628E 01              0.41762E-05              0.63761E 06
  82             0.26630E 01              0.30676E-05              0.86809E 06
  84             0.26631E 01              0.22534E-05              0.11819E 07
  86             0.26633E 01              0.16552E-05              0.16090E 07
  88             0.26634E 01              0.12158E-05              0.21905E 07
  90             0.26634E 01              0.89310E-06              0.29822E 07
  92             0.26635E 01              0.65603E-06              0.40600E 07
  94             0.26636E 01              0.48189E-06              0.55273E 07
  96             0.26636E 01              0.35397E-06              0.75249E 07
  98             0.26637E 01              0.26001E-06              0.10244E 08
 100             0.26637E 01              0.19099E-06              0.13947E 08
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****************
 H=     90.000      B=      0.100
 (PHO)UPPER=      0.020      (PHO)LOWER=      0.098
 DIST1=    60.020      DIST2=    59.778
 (YT/YO)INF=           0.58997E 01      (YB/YO)INF=           0.00000E 00
 C1=                  0.272            C2=                   0.272
 (PHO)LOWER/FEED=      0.831      FEED=      0.118


     N                  YT/YO                      YB/YO                      SF

     1               0.10000E 01                0.81818E 00                0.12222E 01
     2               0.11539E 01                0.70123E 00                0.16455E 01
     4               0.14475E 01                0.51509E 00                0.28102E 01
     6               0.17233E 01                0.37836E 00                0.45547E 01
     8               0.19824E 01                0.27793E 00                0.71328E 01
    10               0.22258E 01                0.20415E 00                0.10902E 02
    12               0.24543E 01                0.14996E 00                0.16366E 02
    14               0.26690E 01                0.11015E 00                0.24230E 02
    16               0.28707E 01                0.80915E-01                0.35478E 02
    18               0.30601E 01                0.59436E-01                0.51486E 02
    20               0.32380E 01                0.43659E-01                0.74167E 02
    22               0.34052E 01                0.32070E-01                0.10618E 03
    24               0.35621E 01                0.23557E-01                0.15121E 03
    26               0.37096E 01                0.17304E-01                0.21438E 03
    28               0.38481E 01                0.12711E-01                0.30274E 03
    30               0.39781E 01                0.93367E-02                0.42608E 03
    32               0.41003E 01                0.68583E-02                0.59786E 03
    34               0.42151E 01                0.50378E-02                0.83670E 03
    36               0.43229E 01                0.37005E-02                0.11682E 04
    38               0.44242E 01                0.27182E-02                0.16276E 04
    40               0.45193E 01                0.19967E-02                0.22634E 04
    42               0.46086E 01                0.14667E-02                0.31422E 04
    44               0.46925E 01                0.10774E-02                0.43556E 04
    46               0.47713E 01                0.79137E-03                0.60292E 04
    48               0.48453E 01                0.58131E-03                0.83353E 04
    50               0.49149E 01                0.42700E-03                0.11510E 05
    52               0.49802E 01                0.31365E-03                0.15878E 05
    54               0.50415E 01                0.23040E-03                0.21882E 05
    56               0.50992E 01                0.16924E-03                0.30130E 05
    58               0.51533E 01                0.12431E-03                0.41454E 05
    60               0.52041E 01                0.91316E-04                0.56990E 05
    62               0.52519E 01                0.67076E-04                0.78297E 05
    64               0.52967E 01                0.49271E-04                0.10750E 06
    66               0.53389E 01                0.36192E-04                0.14751E 06
    68               0.53784E 01                0.26585E-04                0.20231E 06
    70               0.54156E 01                0.19528E-04                0.27732E 06
    72               0.54505E 01                0.14345E-04                0.37997E 06
    74               0.54833E 01                0.10537E-04                0.52039E 06
    76               0.55141E 01                0.77399E-05                0.71243E 06
    78               0.55431E 01                0.56854E-05                0.97497E 06
    80               0.55702E 01                0.41762E-05                0.13338E 07
    82               0.55958E 01                0.30676E-05                0.18241E 07
    84               0.56197E 01                0.22534E-05                0.24939E 07
    86               0.56423E 01                0.16552E-05                0.34088E 07
    88               0.56634E 01                0.12158E-05                0.46580E 07
    90               0.56833E 01                0.89310E-06                0.63636E 07
    92               0.57019E 01                0.65603E-06                0.86916E 07
    94               0.57195E 01                0.48189E-06                0.11869E 08
    96               0.57359E 01                0.35397E-06                0.16204E 08
    98               0.57514E 01                0.26001E-06                0.22120E 08
   100               0.57659E 01                0.19099E-06                0.30189E 08
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