Tendency Modelling Of Desublimator For Optimization And Control

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Abstract

The study was devoted to test the effect of the process variables on the desublimator (cold trap) efficiency under vacuum technique. Since several intermediate process parameters could not be measured or inspected, then it was necessary to predict them by the gray model. Although the novel approach (tendency model) for the desublimator was less accurate than the detailed one, but it showed agreement results when compared with the experimental data. The accuracy of model could be increased by increasing the process variable monitors. The tendency model was more effective to deal with the lack of knowledge about the process when implemented with the optimal control system.

الخلاصة

يتضمن البحث دراسة واختبار تأثير متغيرات العملية على كفاءة المصيدة الباردة بظروف الضغط الفراغي . لمعرفة سلوكية بعض العوامل التي لم يتم قياسها عملياً فقد تـم اقتـراح موديـل رياضـي متخصص هدفه احتساب هذه العوامل بصورة تقريبية . بالرغم من قلة دقة هذا الموديل مقارنة بالموديل النظري التفصيلي إلا انه اعطى نتائج مرضية مقارنة باالنتائج العملية . من الممكن زيادة دقة الموديـل وذلك بزيادة أجهزة القياس للعوامل والمتغيرات المؤثرة على العملية . ان هذا الموديل هو الأفضل عند استخدامه مع مسيطرات متقدمة تحتاج الى معرفة كافية بطبيعة العملية .

Key-word: Desublimator , Tendency model, optimization, control

Nomenclature

a: Constant inherent to each substance
A: Surface area (m^2)
b: Solid layer thickness (m)
c: Molar density of vapour – gas mixture (gmmole/m^3)
Cm: Specific heat of vapour – gas mixture (J / gm.K)
Dv: Binary diffusivity of the vapour – gas mixture (m^2 / sec)
hi: Inside bulk heat transfer coefficient (J / m^2.sec.K)
k: Thermal conductivity of the vapour – gas mixture (J / m. sec. K)
ks: Thermal conductivity of the solid layer (J / m. sec. K)
Mm: Mean molecular weight (gm / gmmole)
M_1: Molecular weight of condensable vapour (gm / gmmole)
Pw: Vapour pressure of desublimated solid on the wall (atm)
P: Total pressure (atm)
Q: Rate of heat generation (Watt)
r: Location from the center of the desublimator (m)
ri: Inside radius of the desublimator (m)
r_o: Outside radius of the desublimator (m)
R: Gas - constant
t: Time (sec)
T: Temperature (C^o)
T_b: Bulk temperature (C^o)

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T_{bath}: Bath temperature (Cº)
Ts: Solid – Layer temperature (Cº)
Tw: Tube wall temperature (Cº)
V: Flow rate of the inert gas (gmmole / sec)
y: Moles of condensable gas per mole of inert gas
Z: Axial direction

Greek letters

ρ: Density of the vapour – gas mixture (gm / m^3)
ρ_s: Density the solid deposit (gm / m^3)
Ø: Angular direction
λ: Latent heat of sublimation (J / gmmole)
α: Heat transfer coefficient (W / m^2.K)

Introduction

Desublimation is encountered in modern chemical technology especially in the catalytic oxidation of aromatic hydrocarbon compounds present in the gaseous phase. On the account of high purity and good appearance of the product, the application of desublimation technique is gained an apparent in the purification of salicylic acid, benzoic acid and naphthalene.

As a method for thermal separation of substances, desublimation technique is competitive with other unit operations such as distillation, crystallization and solvent extraction. Certain inherent characteristics make it unique and in some cases give it a distinct advantage over other separation methods.

The desublimation system is capable of degree efficiency and solids purities higher than 99 %.
(U.S Patent, 2007)

Desublimation technique can be classified in accordance to the applied pressure (wintermantel etal, 1987) into:

i. Vacuum desublimation, where the system pressure is lowered by evacuation as in freeze drying process.
ii. Entrainer desublimation, the partial pressure of a substance is lowered below the pressure of the triple point by controlled addition of an entrainer gas.

The medium where the desublimation process takes place is called "Desublimator"

The specific property of vacuum technique is the production of compact desublimated solid layers which have a density equal five times the density obtained from applying entrainer desublimation technique (Schwenk and Raouzeous, 1995).

The efficiency of the desublimator is affected by many variables, and therefore, determination of these variables and others parameters relate with the desublimation process will be the most important task for the design and control of a desublimator.

It is recommended to use advanced control system to get high efficiency at optimum operating conditions of a desublimator. Since several process parameters can not easily measured, so that the approach (tendency) model for the process now become more desirable to know the path of the process.

Tendency Model

The common approach to design a mathematical model of a process system requires the knowledge of physical and chemical phenomena (such as heat transfer, stoichiometry … etc) to provide detailed mechanism

This technique requires significant effort in identification of many unknown parameters resulting from a complex set of equations. The alternative method

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consists of empirically fitting experimental data; the model represents the system only at specific operating conditions since it may be unrelated to the actual process mechanism. The "tendency" model approach is a compromise between the detailed mechanism and a totally empirical model. Although this technique is usually less accurate than the detailed mechanism, it employs engineering insight into the proposed structure, whose parameters are calculated using available experimental data. Further more, since simulation of this model usually requires less computational time, real time implementation for optimal control is possible.

(Flippi etal, 1989) and (Cawthon and Knaebel, 1989) applied the tendency model approach to chemical reactors, they concluded that, even through the proposed tendency (gray) model might not be as accurate as the true detailed kinetic model, they were expected to successfully guide the process to a more optimal operation.

At the same time they might provide important hints a bout the true kinetics of the process.

From previous works, one can conclude (at this time), the tendency approach almost applied for the chemical reactors.

In the present work, this model was applied to the desublimation process (cold trap) to estimate the unmeasured parameters.

Experimental work

The experimental rig (Figure 1) was designed and constructed into the best way to simulate the real process and to collect the desirable data. The process system consists mainly; mixing chamber, desublimator and vacuum system.

The present work is interested with vacuum desublimation using Benzene-Nitrogen mixture. The monitored operating conditions of the system were:

1. Mole fraction of Benzene vapour at the range of (0.2 to 0.9).
2. Flow rate of inlet mixture from 50 to 250 (cm^3 / min).
3. Desublimator temperature from (- 6 to 2 °C) .
4. Desublimator pressure at the range of (40 to 190 mbar).

The experimental procedure included the following major steps:

1. purging the system by dry nitrogen at pressure of (1200 mbar) and then evacuated to (0.1mbar) for several times until the system to be considered cleaned from any contaminants.
2. Cooling the desublimator to temperature of (- 6 C^o).
3. Entering the feed of (benzene/nitrogen) mixture at desired composition to the evacuated desublimator (at 0.1 mbar).
4. Recording the process variable (pressure, temperature and flow rate) with time by digital sensore.
5. After reaching to equilibrium condition, the noncondensible gas and vapour would be evacuated from the desublimator at constant temperature (-6C^o) and then isolated the cold trap by manual valves.
6. Heating the cold trap to (10 C^o) and drained the liquid benzene to standard bottle.
7. Weighting the liquid benzene into standard bottle.

The samples of gases mixture would be drawn periodically from various locations in the system and then analysised by advanced gas – chromatograph.

The objective function of the process was the efficiency of the desublimator which was:

$$\% \text{ efficiency} = \frac{\text{weight of desublimated solid}}{\text{weight of inlet vapour}} \times 100 \qquad (1)$$

The important parameters which were affected on the desublimated solid but could not be measured were; the thickness and surface temperature of the solid layer. These parameters could be estimated by the tendency model.

The effect of process variables

Four process variables to be considered which were the most effective on the efficiency of the desublimator which were; mole fraction of vapour, flow rate of inlet mixture, pressure and temperature of the desublimator.

The four variables were tested and explained in table (1), which show that the desublimator bulk temperature had proved to be the most effective variable and the pressure inside the desublimator was the most critical variable (Alwan etal, 2000). Also highly interaction could be occurred between these variables then the efficiency had been affected by this interaction.

Optimum conditions

In order to optimize the proper process conditions required for the highest desublimation efficiency, a numerical optimization of the four variables has been achieved through the used of "Modified Hooke and Jeeves" method with equality constraints (Bunday, 1985).

By application the second order regression analysis for the present experimented data, the objective function of the system would be:

$$Y = 81.468 + 1.022 X_1 - 1.653 X_2 - 2.203 X_3 - 0.634 X_4 + 0.099 X_1^2 + 0.099 X_2^2 + 0.099 X_3^2 + 0.099 X_4^2 + 0.005 X_1 X_2 + 0.005 X_1 X_3 + 0.014 X_1 X_4 + 0.005 X_2 X_3 - 0.005 X_2 X_4 - 0.005 X_3 X_4 \quad (2)$$

With constraints of $-2 < X_j < 2$

For j = 1, 2, 3 and 4

Where:

X_1: code level of mole fraction

$$\left(\frac{mole\ fraction - 0.55}{0.175}\right).$$

X_2: code level of flow rate

$$\left(\frac{flow\,rate - 150}{50}\right).$$

X_3: code level of cold trap temperature

$$\left(\frac{Temperature - (-2)}{2}\right).$$

X_4: code level of cold trap pressure

$$\left(\frac{Pressure - 115}{37.5}\right).$$

Y : objective function (efficiency).

The coded levels for each variable had taken between (-2) and (+2) accordance with the central composite rotatable design, suggested by (Cochran, 1957).

The solution of the problem would be obtained with the aid of the computer program (figure 2).

The optimum results (table 2) had shown that the maximum value of desublimation efficiency was obtained when working at a the highest value of vapour mole fraction, lowest value for, inlet stream flow rate, desublimator temperature and pressure.

The predicted model

The prediction of thickness from the developed model was based on the applied experimental conditions, as well as on the calculation of physical properties related to the process vapour-gas mixture existing inside the desublimator. Then the predicated temperature gradient of the solid layer surface could be estimated.

The form of the homogenous solid layer and the distribution of temperatures from the bulk of mixture to the bulk of the cooling bath is shown in figure (3).

Model assumptions

1. Approximately homogenous compact circular and cylinderical solid layer without any vacancies in the structure.

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2. The heat transfer resistance of the tube wall could be neglected because of a thin wall.
3. The resistance through the gas sub-layer near the solid layer to heat and mass transfer was small; therefore it could be neglected.
4. The temperature of the desublimated solid on the wall was assumed to be equal to the wall temperature.
5. The heat and mass transfer processes inside the desublimator would be considered as a quasi steady state process.
6. The inlet mixture to the cold trap was saturated and homogenous, composed of inert gas (nitrogen) and saturated vapour.
7. The heat transfer rate approached a steady state even while frost continued to accumulate.
8. All condensate settles out at the point of formation.
9. The heat and mass transfer processes in the axial direction were very small compared with the transfer in the redial direction; therefore, assumed negligible.

Solid layer surface temperature (TS)

The general thermal equation for the circular and cylindrical coordinate, is shown as follows (James, 1985):

$$\frac{\partial^2 T}{\partial r^2}+\frac{1}{r}\frac{\partial T}{\partial r}+\frac{1}{r2}\frac{\partial^2 T}{\partial \phi^2}+\frac{\partial^2 T}{\partial z^2}+\frac{Q}{K}=\frac{1}{\alpha}\frac{\partial T}{\partial t} \quad (3)$$

For the case of radial transfer, quasi steady state and no generation, equation (3) will be reduced to ordinary differential equation in the following form:

$$\frac{d^2 T}{dr^2}+\frac{1}{r}\frac{d_T}{d\,r}=0 \quad (4)$$

Equation (4) can be integrated twice to:

$$T=C_1 \ln r+C_2 \quad (5)$$

The evaluation of the constant (C_1 and C_2) be the application of the process boundary conditions will produce the following form:

$$Ts=Tw-\frac{T_b-T_w}{In\left(\frac{r_o}{r_o-b}\right)+\frac{k_s}{h_i\left(r_o-b\right)}} In\left(\frac{r_o-b}{r_o}\right) \quad (6)$$

Therefore, Ts can be predicted from equation (6).

Solid layer thickness (*b*)

The rate of condensate deposit per unit area per mole could be determined from a mass balance in accordance with the approach given by (Thompson, 1949). Assuming all condensate settles out at the point of formation. The mass balance can be shown as follows:

$$-V\frac{dy}{dA}=\frac{\rho_s}{M_1}\cdot\frac{db}{dt} \quad (7)$$

Equation (6) could be rearranged in the following form by introducing the term ($hi\,/\,hi$):

$$\frac{db}{dt}=\frac{h_i m_1}{\rho_s}\left(-\frac{V}{h_i}\frac{dy}{dA}\right) \quad (8)$$

By estimate the quantity $\left(-\frac{V}{h_i}\frac{dy}{dA}\right)$ for each point in the desublimator as a function of time, the total deposit thickness at any point could be predicted from the following equation:

$$b=\frac{h_i\,m_1}{\rho_s}\cdot\frac{\lambda.y.\left(T_b-T_w+a_1\right)t}{R.T_b^2.M_m.C_m\left(1+\frac{a_1}{a_2}\right)} \quad (9)$$

Where:

$$a_1 = \frac{\lambda}{M_m . C_m}\left(\frac{C_m . \rho . Dv}{K}\right)^{\frac{2}{3}} . (1-B) . In(1+y)$$

$$a_2 = \frac{R . T^2}{\lambda . Y}\left(\frac{C_m . \rho . Dv}{K}\right)^{\frac{2}{3}} . (1-B) . In(1+y)$$

$$\beta = \frac{Pw}{P}$$

The proposed structure of the tendency model was obtained by the simulation of equations (1,6 and 9) with the aid of the MATLAB computer program.

The application of the tendency model for the present desublimator yields agreement results for thickness of the solid layer when compared with that experimentally measured (by the portable type ultrasonic detector) as shown in figure (4), with standard deviation of 0.27. This result could be attributed to the tearing of the desublimated solid particles from the surface of solid layer which occurred due to the forced stream inside the desublimator which was not taken into account by the approach model.

Also agreement results were obtained when compared the experimental and predicted efficiency as a function of temperature as shown in figure (5) with standard deviation of 0.08. This indicates that the highest efficiency is obtained when working at the lowest value of desublimator temperature in the experimental range used. The experimental and predicted efficiency were estimated regarding to equation (1). The accuracy of the tendency model can be increased by increasing the monitors of the process variables.

The proposed computerized-control system

As mentioned, since the effective process variables which were highly interacted and affected on the efficiency of the desublimtor, so that the on-line multiple PID control with non-interacting loops must be needed to operate the system at optimum conditions (Borrie,1986). The tendency model is accurate to deal with the lack of knowledge about the process parameters. Simulation of this model for digital control is required to less computional time compared with the detailed model, also the real time implementation for the adaptive control algorithm will be more effective. Figure (6) explains the computer control function which operated by the aid of the predicated tendency model.

Conclusions

1. The objective function of the desublimator was the efficiency of the vapour trap, which was affected by four process variable these were; vapour mole fraction inlet mixture flow rate, desublimator temperature and pressure. The temperature had proven to be the most effective variable while the pressure was the most critical variable for the vacuum desublimation technique.
2. Although the tendency model was less accurate than the detailed model, but it showed agreement results when applied to desublimator.
3. Simulation of tendency model for computer control was required to less computional time compared with the complex detailed model of cold trap.
4. Model predictive control (adaptive) required an accurate model of the process which was often not available. To deal with this lack of knowledge about the process, an effective (tendency) model was utilized.

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Table (1): Effect of the process variables on the desublimator – efficiency.

Mole fraction	Flowrate Cm^3 / min	Temperature °C	Pressure mbar	Efficiency %
0.55	50.0	-2	115.0	85
0.55	150.0	-6	115.0	88
0.55	150.0	-2	40.0	84
0.9	50.0	-6	3.0	99.5

Table (2): Results of the search for the maximum by Hook and Jeeves method.

Search No.	X_1	X_2	X_3	X_4	Y
Starting	- 2.0	- 2.0	- 2.0	- 2.0	90.064
	- 1.8	- 2.0	- 2.0	- 2.0	90.180
	- 1.8	- 1.8	- 2.0	- 2.0	89.775
	- 1.8	- 2.0	- 1.8	- 2.0	89.665
	- 1.8	- 2.0	- 2.0	- 1.8	89.444
	- 1.9	- 2.0	- 2.0	- 2.0	90.121
	----------	----------	----------	----------	----------
1	- 1.8	- 2.0	- 2.0	- 2.0	90.180
	----------	----------	----------	----------	----------
2	- 1.6	- 2.0	- 2.0	- 2.0	90.311
	----------	----------	----------	----------	----------
3	- 1.4	- 2.0	- 2.0	- 2.0	90.440
	----------	----------	----------	----------	----------
4	- 1.2	- 2.0	- 2.0	- 2.0	90.589
	----------	----------	----------	----------	----------
5	- 1.0	- 2.0	- 2.0	- 2.0	90.730
	----------	----------	----------	----------	----------
6	- 0.8	- 2.0	- 2.0	- 2.0	90.900
⋮	----------	----------	----------	----------	----------
⋮	----------	----------	----------	----------	----------
⋮	0.0	- 2.0	- 2.0	- 2.0	91.616
⋮	----------	----------	----------	----------	----------
⋮	----------	----------	----------	----------	----------
⋮	0.4	- 2.0	- 2.0	- 2.0	92.021
⋮	----------	----------	----------	----------	----------
⋮	----------	----------	----------	----------	----------
20 (Final)	2.0	- 2.0	- 2.0	- 2.0	93.960

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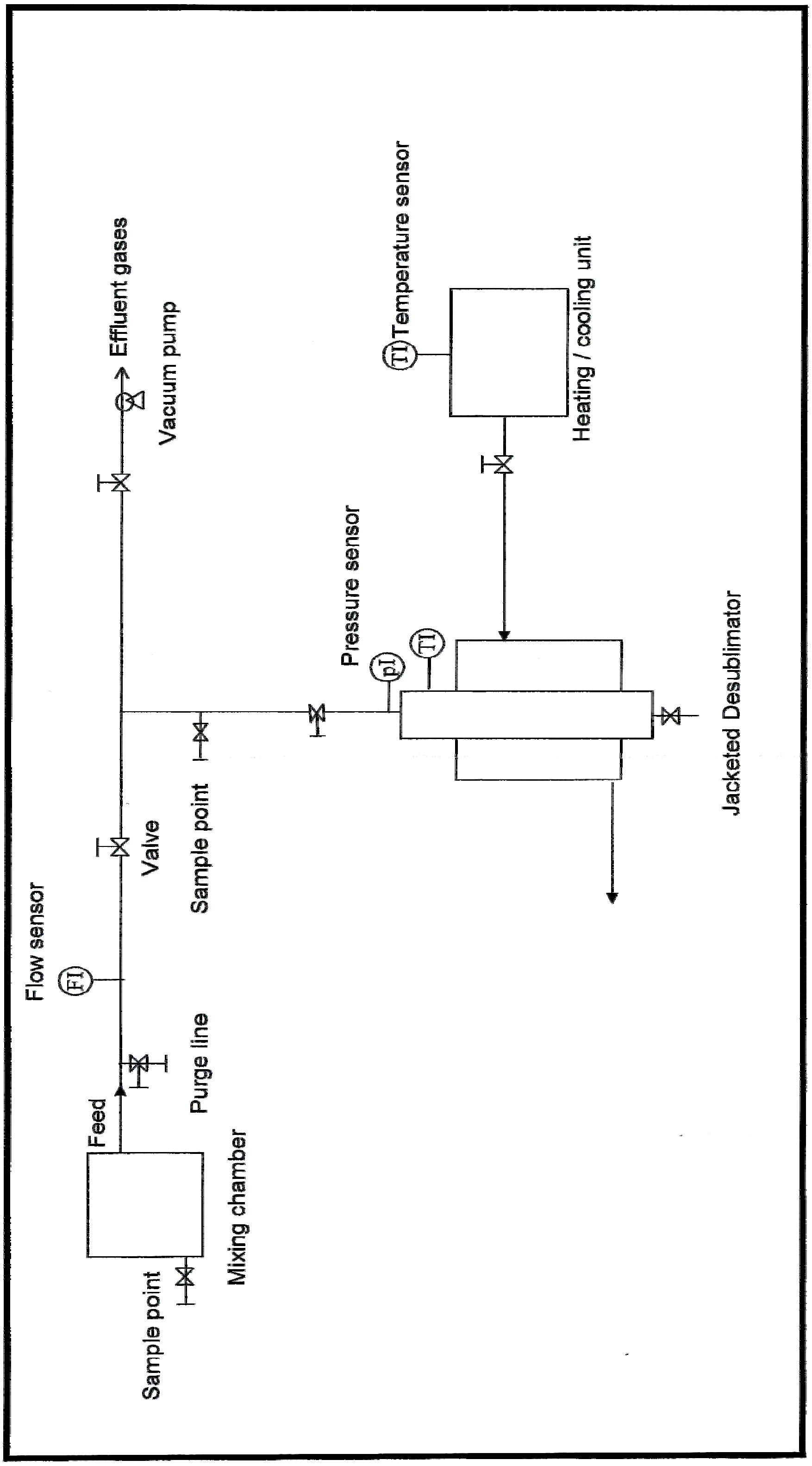
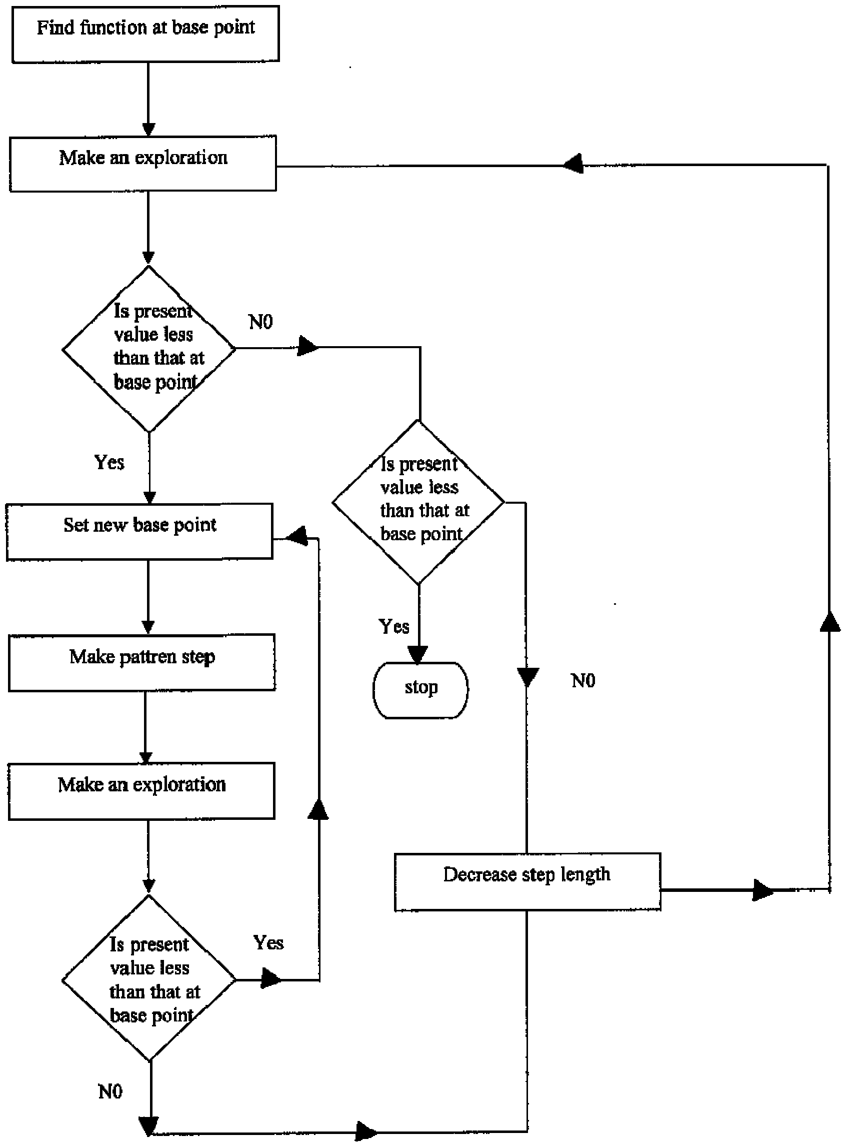


Figure (1): Diagram of experimental set-up.


Find function at base point
Make an exploration
Is present value less than that at base point
N0
Yes
Set new base point
Is present value less than that at base point
Yes
stop
N0
Make pattren step
Make an exploration
Decrease step length
Is present value less than that at base point
Yes
N0

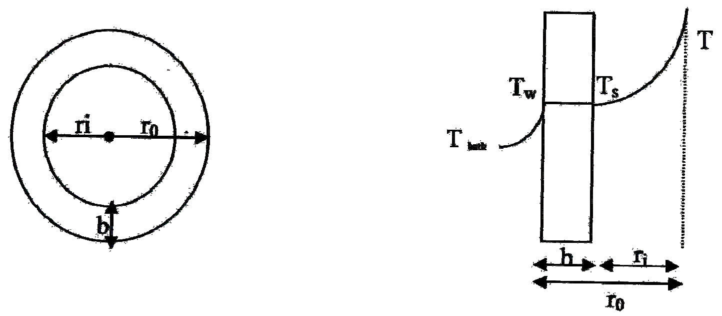


Figure (3): Temperature distribution between the inside bulk of the desublimator and the cooling bath.

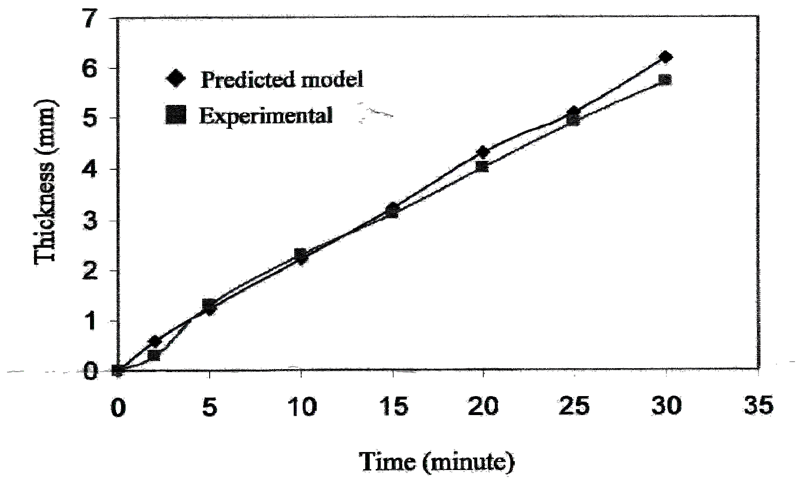


Figure (4): The Comparison between the Predicted and Experimental solid layer thickness.

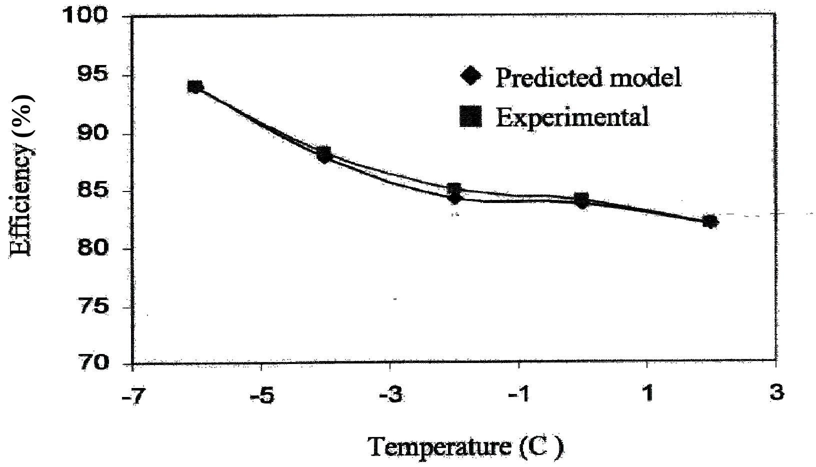


Figure (5): The Comparison between the Predicted and Experimental efficiency.

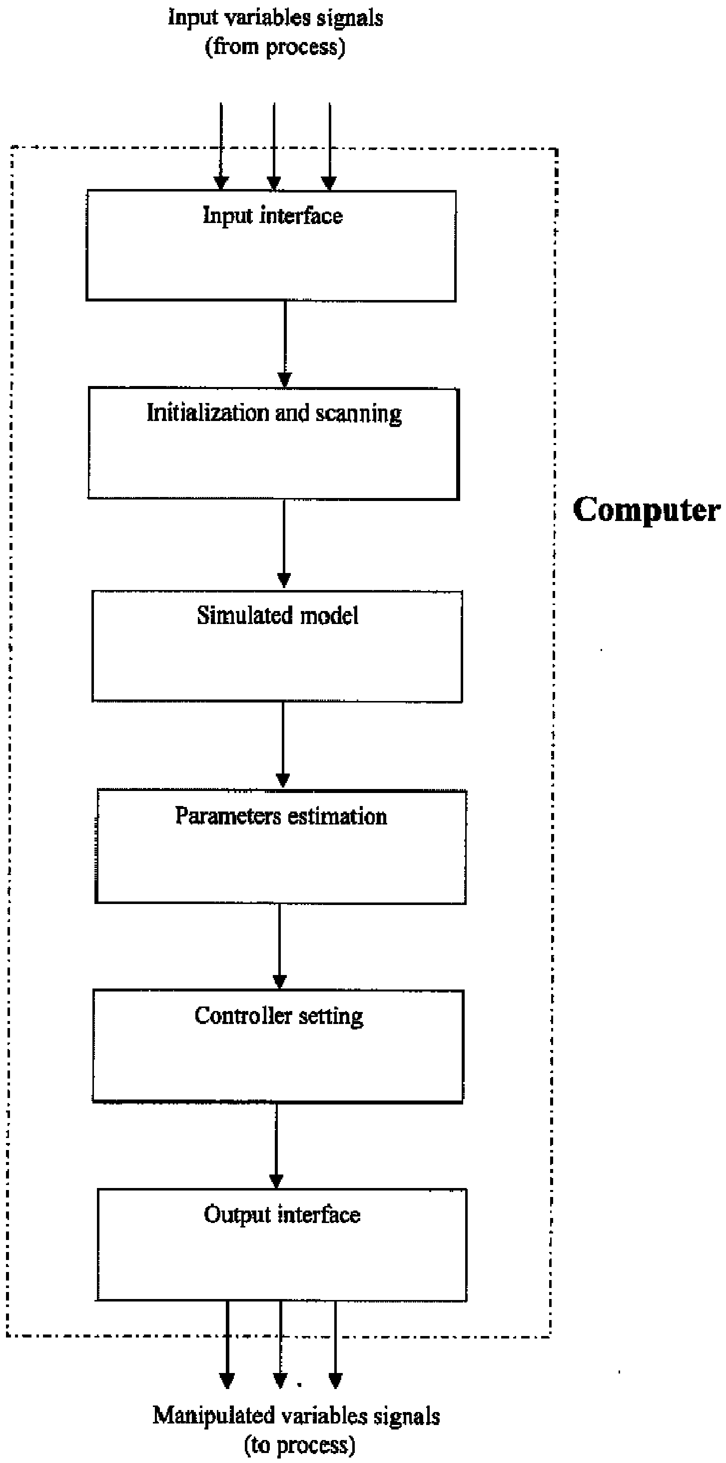


Figure (6): Block diagram of the computer control function.