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Simulation and Modeling of an Integrated Process Route for the Synthesis of Vinyl Chloride Monomer from Acetylene: Factorial Design Method and Artificial Neural Network

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ABSTRACT

Vinyl Chloride gas is a nonirritating and colorless substance. It is usually colorless at a concentration lower than 3900 ppm (10,000 mg/m3). Vinyl Chloride is simply compressed to liquid for storage and shipping. At a concentration between 200 and 500 mg/m3, a Sweetish odor of Vinyl Chloride may be detected. This research paper is focused on the simulation of an integrated process route for the synthesis of Vinyl chloride Monomer from Acetylene via Aspen Hysys Simulation as well as the Factorial Design of the experiment with MINITAB 17.0. Fit Regression and Artificial Neural Network were employed for the modeling of the responses. Molar flow rates of acetylene (C2H2) and hydrogen chloride (HCl) predicts the conversions of acetylene and hydrogen chloride. A recycle unit is added to the process flow diagram and the maximum conversion of C2H2 and HCl is found to be 99.90 and 99.80 %, respectively. Analysis of variance (ANOVA) gives the results of the statistical correlation between the independent variables and response variables. The simulation and modeling results reveal that the Artificial Neural Network model gives better prediction and analysis of the process route with correlation coefficient (R squared values) of 97.921 % and 98.423 % for the conversion of C2H2 and conversion of HCl, respectively compared to the Factorial Design Method model with R squared values value of 79.47 % and 73.70 % for the conversion of C2H2 and conversion of HCl, respectively.

Keywords: *Vinyl chloride Monomer, Conversion, Aspen Hysys simulation, Factorial Design Method, Artificial Neural Network.*

1.0 INTRODUCTION

1.1 Vinyl Chloride

Vinyl chloride (VC) is a nonirritating, colourless gas at standard temperature and pressure. It is generally odorless below 10 000 mg/m^3 (3900 ppm), but a sweetish odour may be detected by some sensitive individuals between 200 and 500 mg/m^3. The gas is liquefied under pressure with ease and is frequently stored or shipped as a liquid [1]. Vinyl chloride is highly stable in the absence of sunlight or oxygen. Above 400 °C, it dissociates into acetylene and hydrochlorine. In the atmosphere, VC reacts with hydroxyl radicals and ozone, ultimately forming formaldehyde, carbon monoxide, hydrochloric acid and formic acid. On the basis of measured reaction rates with hydroxyl radicals and their concentration in air, it is estimated that the half-time of VC in the atmosphere is about 20 hours [2]. Vinyl chloride, essentially all of which is used to synthesize poly (vinyl chloride) and vinyl chloride copolymers, is the chief tonnage organochlorine compound produced, with the exclusion of its precursor, ethylene dichloride. Vinyl chloride monomer (VCM) finds its chief sources in order of significance from VC production plants, polyvinyl chloride (PVC) polymerization infrastructures, and plants where PVC products are fabricated. Minor sources include storage and handling facilities for VC and PVC and plants producing ethylene diamine or ethylene dichloride. VC emissions have been reported from municipal landfills in the United States, but the precise source of emission is uncertain and

systematic survey data are unobtainable [1]. Vinyl chloride monomer finds good applications in medical equipment, building materials, packaging materials, electronics equipment, etc. Roughly 5 million tonnes of VC were produced in the entire region of Europe in 1981. The levels of emission from VC and PVC production plants and facilities depend upon the processes and control technology engaged. The implementation of the best existing technology can reduce emissions below 1% of production volume, nevertheless emissions from facilities in some nations surpass this value [2]. For the acetylene-based process, Vinyl chloride has been prepared since the early 1930s by the reaction of hydrogen chloride with acetylene is presented in (1) [3]:

$$CH \equiv CH_{(g)} + HCl_{(g)} \longrightarrow CH_{2(g)} = CHCl_{(g)} \qquad (1)$$

The reaction occurred in the vapor phase, in the presence of a catalyst containing mercuric chloride supported on active carbon, at temperatures between 97 and 177 ^{0}C depending on the age and activity of the catalyst. Due to ageing of the catalyst, it activity drops; and to sustain the reaction rate, the temperature is elevated [3]. The yields obtained from this process is within 80% to 95%, with a by-product is 1,1-dichloroethane. The process is highly desirable as the reaction conditions are mild, it is a one-stage process, the stoichiometry is favorable and heat is not supplied since the reaction process is exothermic [3].

1.2 Factorial Method, Artificial Neural Network And Simulation

Factorial and fractional factorial designs are usually used as experimental strategies to evaluate the impact of several factors on a response process variable [4]. Total randomization of the experimental runs is often not practical when it is hard to alter the levels of some of the factors [4]. Factorial design experiments include concurrently more than one factor and each factor is at a minimum of two levels. Numerous variables or factors have simultaneous effect on the behavior under investigation in factorial design experiments. Hence, the alchemist is concerned with the main and interaction effects amongst dissimilar and independent variables or factors. Factorial experiment is said to be "symmetrical" if the number of levels for all factors are the same, and "mixed" if the number of levels of each of the factor are different. Considering the significance of factorial design methods, they are said to be more effective than experiments with one-factor-at-a-time and it is essential in presence of interactions to circumvent deceptive inferences. Factorial design methods also allow the influence of an independent variable to be projected at numerous levels of the other independent variables, giving rise to valid deductions over a variety of experimental conditions [4].

Artificial Neural Network (ANN) has over the years made a remarkable contribution to various fields including engineering and technology. As such, it has brought about advancement in the fields [5]. Artificial neural network is a modelling technique which simulates the operation of the brain and nervous system [6]. It is an information processing system that does specific function similar to the biological neural network [7]. In this pattern, the main element is the innovative structure of the information processing system that is composed of highly interconnected processing element working in unison to solve a specific problem. Artificial Neural Networks learn by example like humans do [8]. Artificial neural network is configured to do a specific work which includes data classification or pattern recognition through the process of learning. In biological systems, learning involves the adjustment of the synaptic connection that exists between the neuron [9]. Artificial neural networks are typically complex communication networks wired together from hundreds of simple processing unit or nodes. Each unit is a model of the neuron which sends off a new signal when it received strong and sufficient input signal from the other nodes which it is connected [8]. Hence, ANN resembles the brain in two ways. Firstly, the network acquires knowledge through the process of learning. Secondly, the knowledge is stored using the interneuron connection strengths which are the synaptic weights [10]. ANNs are applicable in quantifying nonlinear relationship between a model dependent and independent variables by means of iterative training of data obtained [11]. This is similar to conventional statistical model where error between model outs and corresponding measured values (training set) of a particular data set are minimized by adjusting the model parameters (such as connection weights) in the model calibration phase [12]. Performance of ANNs can be improved by developing the ANN models in a systematic manner which will address major factors, including the determination of adequate model inputs, data division and pre-processing, the choice of a suitable network architecture, careful selection of some internal parameters that control the optimization method, the stopping criteria and validation of model [13].

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Process simulation defines the imitation of a real-time process that represents its operation over time. Simulation is often used with scientific modeling of chemical engineering processes or systems to give an insight of the behavior of the particular process or system. It displays the eventual real effects of substitute conditions and courses of action. Computer simulation of a chemical process models the process in a way it emulates a real-time process, usually on a pilot scale. Aspen HYSYS is a software package for computer simulation of chemical processes and useful in industries such as chemical, process optimization, bio-chemical, process control, wastewater treatment, pharmaceutical, petrochemical, oil and gas, etc. [14], [15].

2.0 METHODOLOGY

Vinyl Chloride Monomer was synthetized by simulating the process route presented in figure 1.0 with Aspen Hysys 8.8. The integrated route consists of addition of a recycle unit for recovering unreacted feed gases for further reaction in order to achieve 99.90 % conversion for Acetylene and 99.98 % conversion for hydrogen chloride from the reaction process, a cooling device for further cooling of the separator product and a column in order to obtain a stream of approximately 99.98 % purity of the Vinyl Chloride Monomer. Factorial Method was used to design the experiment; Fit Regression of linear model and Artificial Neural Network were used to model and predict the responses (Hysys Simulation results).

2.1 Process Route Description

The integrated computer-simulated process for the synthesis of Vinyl Chloride from Acetylene is presented in figure 1.0. Within the process route, the reactant gases are mixed in a Feed mixer (B1) and then Compressed in (B1) for better interaction of the reactant gases. The compressed gases are fed into a Process conversion reactor (B3) where Acetylene is chosen as the base component and setting the default conversion to 90 %. The reactor top exit stream is cooled in a box cooler (B4) and Short column (B5) with top product recycled back to the chest of the reactor to achieve 99.90 % conversion for acetylene and 99.89 % conversion for hydrogen chloride. The bottom product is the VCM rich stream which exits at a low temperature of -16.62 ^{0}C and heated in a heater (B6) in order to elevate the temperature to 25 ^{0}C. The heat of reaction depicts that the reaction process is exothermic with the value -2.4 x 10^{4} kcal/kgmole. The exit stream from B6 is further sent to a component splitter (B7) to further separate the VCM monomer from other components (substances), hence achieving a mole composition of 1.00 (100 %) in the pure VCM stream. The equipment design parameter and experimental design values presented in tables 1.0 and 2.0, respectively.

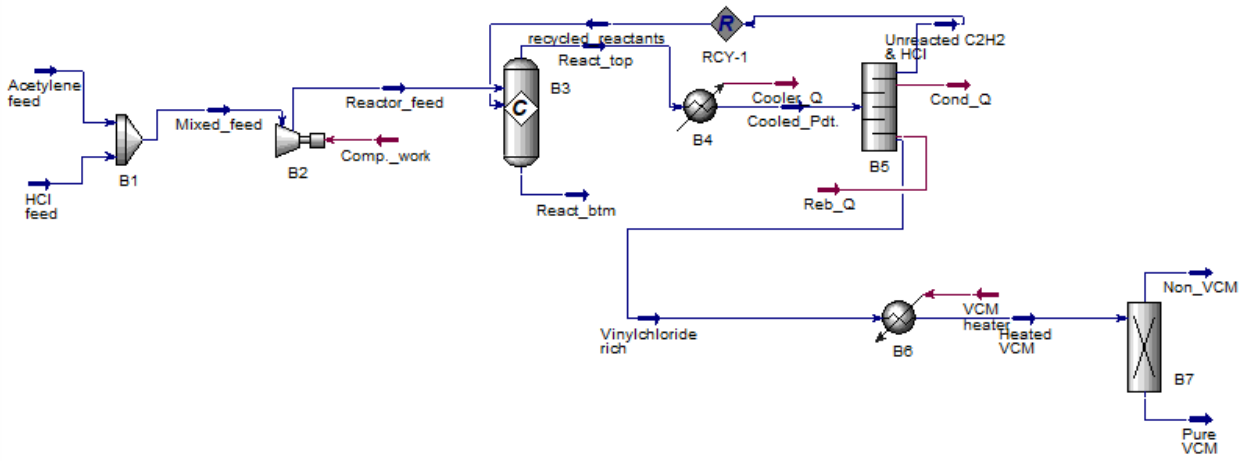

Figure 1.0: Integrated Computer-Simulated PFD for the synthesis of VCM from Acetylene

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Table 1.0: Equipment design parameter and value

Feed Mixer (B2)	elevation (Base) (m)	elevation (Ground) (m)	Diameter (m)				
	1.00	2.00	2.00				
Compressor (B2)	Adiabatic head (m)	Polytropic head (m)	Adiabatic eff. (%)	Polytropic eff. (%)	Duty (kW)	Capacity (ACT-m^3/h)	Delta P (bar)
	1.300×10^4	1.330×10^4	85.00	86.971	195.2 kW	3695	3.00
Process Reactor (B3)	Delta P (bar)	Vessel volume(m^3)	Liquid level (%)	Liquid volume (m^3)	Reaction heat (kcal/kgmole)		
	0.0000	300	50.00	150.00	-2.4×10^4		
Top Product Cooler (B4)	Delta P (bar)	Delta T (^{0}C)	Duty (kcal/h)				
	0.00	-1027	1.865×10^6				
Product Separator (B5)	Condenser pressure (bar)	Reboiler pressure (bar)	External Reflux ratio	Internal Reflux ratio	Min. N0. of Trays	Max. N0. of Trays	Optima feed stage
	1.00	1.00	3.50	2.530	4.278	8.549	5.849
VCM rich Heater (B6)	Delta P (bar)	Delta T (^{0}C)	Duty (kcal/h)				
	0.00	41.60	4.157×10^5				
Pure VCM Splitter	Products	Vapour Fraction	Temperature (^{0}C)	Enthalpy	Pressure (kPa)		
	Non VCM	1.000	30.00	1.587×10^4	150.0		
	Pure VCM	1.000	30.00	8.375×10^3	311.1		
RCY-1	Maximum iteration	Flash Type	Acceleration Frequency	Q Maximum	Q Minimum	Acceleration Delay	
	10	PT Flash	3	0.00	-20.00	2	

Table 2.0: Experimental Design

Levels	-1	0	1
Flow rate of acetylene (A) (kgmole/hr)	50	75	100
Flow rate of hydrogen chloride B (kgmole/hr)	50	75	100

3.0 RESULTS AND DISCUSSION

Experimental design was done using Factorial Method with two independent variables, each having three levels; and two response variables. The independent variables are the flow rates of acetylene and HCl, while the response variables are the conversion of acetylene and HCl. The factorial design method gives nine (9) experimental (simulation) runs for the simulation of the process, and the results obtained from the computer simulation of the integrated process route are shown in table 3.0:

Table 3.0: Hysys Simulation Results

Flow rate of Acetylene	Flow rate of HCl	Conversion of Acetylene	Conversion of HCl
0.0000	0.0000	90.0901	90.0996
0.0000	1.0000	90.0905	67.854
-1.0000	0.0000	90.0906	60.4425
1.0000	0.0000	75.0751	100
-1.0000	1.0000	90.0912	45.6163
0.0000	-1.0000	66.7333	100
1.0000	-1.0000	50.0500	100

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1.0000	1.0000	90.0902	90.095
-1.0000	-1.0000	90.0902	90.0996

3.1 Statistical Analysis of The Factorial Method

Factorial method was used to model the relations between the different simulation variables and their outcome on the conversion of the reactants, based on the "Fit Regression Model" of MINITAB 17.0 (Pen, USA). The response variable was modeled through a polynomial model and analysis of variance (ANOVA) was done to assess the implication of each independent variable on conversion. Multiple regression analysis was used to develop a mathematical model that relates the simulated response to the independent variables. A general description for the Response is given by the polynomial model presented in (2):

$$Y = \alpha_0 + \sum_{i=1}^{n} \alpha_i X_i + \sum_{i=1}^{n} \alpha_{ii} X_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \alpha_{ij} X_i X_j + \varepsilon \qquad (2)$$

Defining the terms in the model, X_1, X_2, X_3, …, X_n are the predicting coded variables, α_0 is the offset term, and α_i accounts for the linear effects, α_{ii} accounts for the squared effects and α_{ij} accounts for the effects. The random error is denoted by ε. A model decrease may be convenient, if there are many redundant terms of the model [17]. The summary of the statistical model based on the Lack-of-Fit Test explicated the fitness of the models. The significance of each variable in the model was assessed using ANOVA and mathematical models were obtained based on the general response model of (2). The models obtained are (3) and (4), and were used for the prediction of the conversion (X) of acetylene and conversion (X) of hydrogen chloride, respectively. In these equations, the flow rates of acetylene and hydrogen chloride are represented as A and B, respectively.

$$X_{C_2H_2}(\%) = 60.8 - 0.278\,A + 0.533\,B \qquad (3)$$

$$X_{HCl}(\%) = 84.3 + 0.537\,A - 0.538\,B \qquad (4)$$

The ANOVA of the linear regression model showed the significant level of the model at 79.47 % and 73.70 % for the conversions of acetylene and hydrogen chloride, respectively (Tables 4.0 and 5.0). This reveals how better the model fits the simulation results. Hence, it infers that the total variance in the response could be described using this mathematical model. Results from regression analysis shows the statistical correlation and prominence between the independent variable and the response variable. R^2 is the correlation coefficient, which describes the percentage of conversion (%) variation that is explained by its relationship with flow rates of acetylene and hydrogen chloride. So, the R^2 (*adjusted*) is percentage of conversion (%) variation that is explained by its relationship with flow rates of acetylene and hydrogen chloride, adjusted for the number of independent variables in the model. This adjustment is vital since R^2 for this model increases when a new predictor is added. Hence, the adjusted R^2 is a valuable tool for comparing the explanatory strength of models with different predictors. The null hypothesis was tested using the P-values of 0.009 and 0.018 for each coefficient and indicates that the coefficient has no effect [17].

Table 4.0: Analysis of Variance for the conversion of Acetylene

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	2	1343.96	671.978	11.62	0.009
Flow rate of Acetylene	1	280.33	280.330	4.85	0.007
Flow rate of HCl	1	838.75	838.749	14.50	0.009
Error	6	347.12	57.854		
Lack-of-Fit	5	347.12	69.425	8.67811×10^8	0.000
Pure Error	1	0.00	0.000		
Total	8	1691.08			
Model Summary					
S	R-sq	R-sq (adj)	R-sq (Pred)		
7.60619	79.47 %	72.63 %	49.30 %		
Coefficients					

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Term	Coef	SE Coef	T-Value	P-Value	VIF
Constant	60.8	15.9	3.82	0.009	
Flow rate of Acetylene	-0.278	0.126	-2.20	0.070	1.04
Flow rate of HCl	0.533	0.140	3.81	0.009	1.04

Table 5.0: Analysis of Variance for the conversion of HCl

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	2	2323.7	1161.84	8.41	0.018
Flow rate of Acetylene	1	1043.1	11043.13	7.55	0.033
Flow rate of HCl	1	853.0	853.00	6.17	0.048
Error	6	829.3	138.22		
Lack-of-Fit	5	389.6	77.91	0.18	0.937
Pure Error	1	439.8	439.77		
Total	8	3153.0			
Model Summary					
S	R-sq	R-sq (adj)	R-sq (Pred)		
11.7568	73.70 %	64.93 %	36.76 %		
Coefficients					
Term	Coef	SE Coef	T-Value	P-Value	VIF
Constant	84.3	24.6	3.43	0.014	
Flow rate of Acetylene	0.537	0.195	2.75	0.033	1.04
Flow rate of HCl	-0.538	0.216	-2.48	0.048	1.04

3.1.1 Graphical Representation of the Mathematical Model

Figures 2.0 and 3.0 show the contour plots for the conversion of acetylene and conversion hydrogen chloride, respectively. This explore the potential relationship between independent variables (flow rate of acetylene & flow rate of hydrogen chloride) and response variables (conversion of acetylene and conversion of hydrogen chloride). Hence, it displays the three-dimensional relationship in two dimensions, with x- and y-factors (predictors) on the x- and y-scales and response values represented by contours. From the legend of these plots, it is observed that maximum conversion of over 90 % of both acetylene and hydrogen chloride is observed in a purple-colored region.

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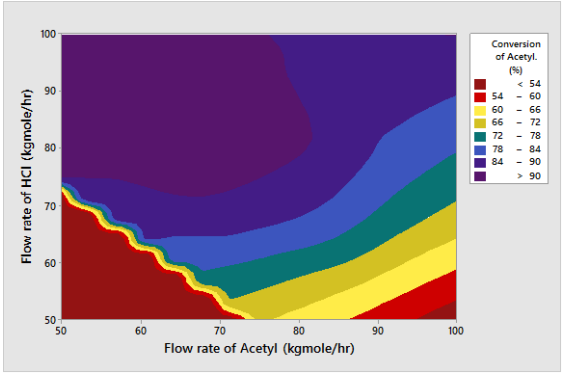

Figure 2.0: Contour plot for the conversion of Acetylene

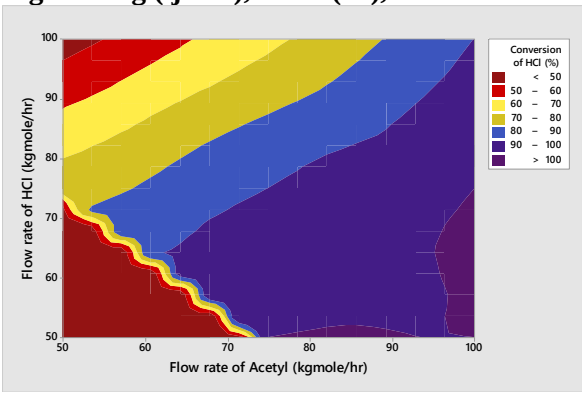

Figure 3.0: Contour plot for the conversion of HCl

Figures 4.0 and 5.0 is the surface plots for the conversion of acetylene and conversion of Hydrogen chloride, respectively. This surface plots are generated from the full factorial design of the experiment. The 3D surface plots also showed how the conversion strongly rely on the flow rates of the reactants.

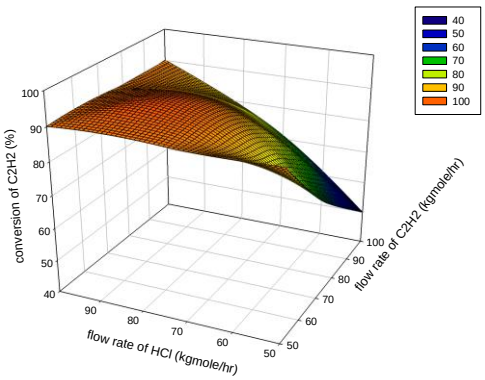

Figure 4.0: Surface plot for the conversion of Acetylene

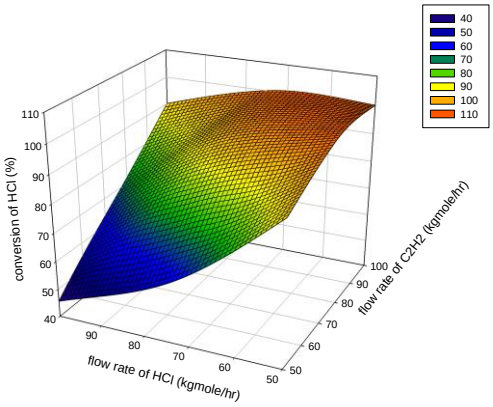

Figure 5.0: Surface plot for the conversion of HCl

3.2 Artificial Neural Network (ANN)

Neural Builder (NB) of Artificial Intelligence Software, Neurosolutions 6.0 was used for the ANN modeling. Within the Neural network training environment, “Generalized feed forward” was adopted as the method of training and “Genetic Algorithm” as the input optimization. A hidden layer of 1 was used and this could train the simulation data better compared to higher values of hidden layers. Momentum value of 0.70 and step size of 0.1 was used as the learning rule, and selecting LinearTanhAxon as the transfer function for the ANN simulation. The Generalized ANN structure with 1 hidden layer for training the Hysys simulation results is presented in figure 6.0.

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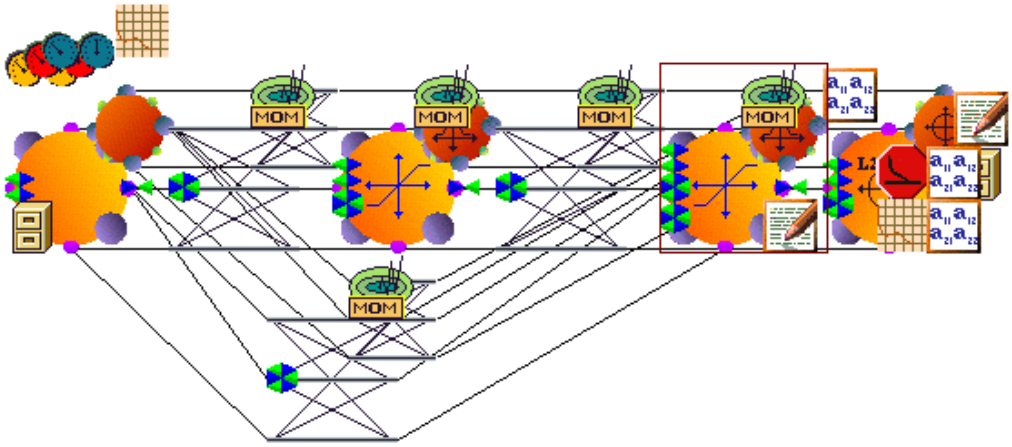

Figure 6.0: The Generalized NeuroBuilder for the ANN simulation

The Neural network simulation is completed at an epoch number of 245 while training the results for the conversion of acetylene and epoch number of 202 while training the results for the conversion of HCl. The active performance for the ANN simulation is shown in table 6.0. The R squared values of the 97.921 % and 98.423 % for X_{C2H2} and X_{HCl}, respectively as shown in ANN active performance results (Table 6.0) indicates the adequacy and better prediction of the ANN model. Also, the low values of the mean square error (MSE), normalized means square error (NMSE) and % Error, indicates a slight variation which could be neglected while predicting the response variable. The "confusion matrix" is also identified as the "error matrix". It permits the visualization of the performance of the ANN algorithm with "true positive" value of 100, "true negative" value of 100, "false positive" value of 0, and "false negative" value of 0,. The Accuracy, Precision and Recall values of the confusion matrix is found to be 1.0. Hence, it defines the performance of the model for which the actual values are known.

Table 6.0: ANN Active Performance

ANN Active Performance for X_{C2H2}			ANN Active Performance for X_{HCl}		
MSE	0.01898		MSE		0.01904
NMSE	0.04998		NMSE		0.04498
R Square	0.97921		R Square		0.98423
% Error	3.90714		% Error		4.94395
AIC	12.3202		AIC		16.3102
MDL	-9.31312		MDL		-6.7364
ANN Active Confusion Matrix					
	Positive	Negative		Positive	Negative
Positive	100.0000	0.0000	Positive	100.0000	0.0000
Negative	0.0000	100.000	Negative	0.0000	100.000

Table 7.0 and 8.0 compares the predicting strength of the factorial method and artificial neural network. Solving the models shows the predicting values of each model. The strength of prediction of the Factorial Method and ANN model is best explained with their coefficient of determination (R squared). The results shows that ANN with R squared values 97.921 % and 98.423 % for X_{C2H2} and X_{HCl}, respectively could give better prediction of the Hysys simulation results compared to the factorial method with R squared values 79.47 % and 73.70 % for X_{C2H2} and X_{HCl}, respectively.

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Table 7.0: Comparison of Factorial Method and ANN for the Conversion of Acetylene

Flow rate of C_2H_2	Flow rate of HCl	Hysys Simulation	Factorial Method	A.N.N modeling
0.0000	0.0000	90.0901	79.925	82.6711
0.0000	1.0000	90.0905	93.25	92.3157
-1.0000	0.0000	90.0906	86.875	92.3157
1.0000	0.0000	75.0751	72.975	67.3939
-1.0000	1.0000	90.0912	100.2	92.3157
0.0000	-1.0000	66.7333	66.6	70.9139
1.0000	-1.0000	50.0500	59.65	59..6925
1.0000	1.0000	90.0902	86.3	92.3157
-1.0000	-1.0000	90.0902	73.55	92.3157
R Squared			79.49 %	97.921 %

Table 8.0: Comparison of Factorial Method and ANN for the Conversion of HCl

Flow rate of C_2H_2	Flow rate of HCl	Hysys Simulation	Factorial Method	A.N.N modeling
0.0000	0.0000	90.0996	84.225	73.1191
0.0000	1.0000	67.854	70.775	74.5215
-1.0000	0.0000	60.4425	70.8	67.2060
1.0000	0.0000	100	97.65	103.0213
-1.0000	1.0000	45.6163	57.35	60.1340
0.0000	-1.0000	100	97.675	93.1416
1.0000	-1.0000	100	111.11	93.3648
1.0000	1.0000	90.095	84.2	86.6509
-1.0000	-1.0000	90.0996	84.25	73.286
R Square			73.70 %	98.423 %

4.0 CONCLUSION

Statistical analysis and modeling using full factorial and the Regression Fit Model of statistical analysis software, MINITAB 17.0 was carried out to assess the conversion of reactants from the simulation of an integrated process flow diagram for the synthesis of vinyl chloride monomer from acetylene with process simulation software, Aspen Hysys 8.8. Artificial Neural Network (ANN) of Neurosolutions 6.0 was also used to train and test the Aspen Hysys simulation results. The effect of molar flow rate of acetylene and HCl was examined while the conversion of the reactants was assessed by Factorial method and Artificial Neural Network. The process response variable (conversion) showed a stochastic distribution with respect to the predicting variables, with the maximum conversion of approximately 90 % and 89 % for acetylene and HCl, respectively before a recycle unit is added; and maximum conversion of 99.90 % and 99.89 % for acetylene and HCl, respectively after a recycle unit is added. sThis increase in the conversion after a recycle reveals that in chemical process, recycle units improves the performance of a process and there by optimize (increase) the results of an expected response. This shows a better and improved performance than the early study of [14] and conforms to the early study of [17]. The research work has revealed that the models used can be recommended for simulation and design of a pilot plant for the synthesis of Vinyl chloride monomer from acetylene. The model results reveals that ANN gives better model for prediction and analysis of the simulated process route with R squared values of 97.921 % and 98.423 % for the conversion of C_2H_2 and conversion of HCl, respectively compared to FM with R squared values of 79.47 % and 73.70 % for the conversion of C_2H_2 and conversion of HCl, respectively.

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