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PART A

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Dynamic behavior of a multi-tasking reactive distillation column for production of silane, dichlorosilane and monochlorosilane

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Abstract

In recent decades, the production of solar cells, along with the associated technologies has become important research topics in the field of chemical engineering, due to the increasing application of solar energy. The solar cell manufacturing is based on solar grade silicon, which can be obtained by decomposition of silane to trichlorosilane. The production cost of this component is highly dependent on the required purity. Proposing a conceptual design of a reactive distillation column (RD) multitasking for producing high purity silane is presented as well as monochlorosilane and dichlorosilane. The latter also being products of great commercial and industrial interest in the energy sector. It also set the objective of scheduling the operating conditions to ensure optimum dynamic performance. It has been shown that it is feasible to obtain a single multitasking design, in which the three products of interest are obtained in the dome of the column, this varying operating conditions such as reflux ratio and reboiler heat duty so that it remains the system under stable conditions. It was possible to obtain purities of 99.5% for all three products and the complete conversion to trichlorosilane silane through a reactive distillation column. In this paper, the properties of said column control were also evaluated. In order to observe the dynamic behavior of the multitasking reactive distillation column, this system is tested under various control strategies: temperature, composition and cascade (temperature/composition). Monitoring results shows that the composition and temperature control structures are the best configurations, as it provides dynamic advantages, but in general with those three control strategies a good dynamic behavior could be observed.

Keywords: dynamic behavior, multi-tasking reactive distillation column, silane, dichlorosilane and monochlorosilane.

1. Introduction

The growing interest in obtaining silicon raw material for solar cells has evolved significantly in recent decades, (Braga et al., , 2008). A more detailed analysis indicates that the photovoltaic market has increased at an average rate of 45% per year over the past decade, showing a major demand between 2007-2011 up to 70% per year; with a decrease of 15% in 2012 because some European countries reduced the incentives for its implementation. Although several reports indicate continuous growing in this sector, the entire capacity installed at 2011 was 27 GW which only represents 1% of the total energy production considering all available sources. New technologies based on solar cells of advanced materials have been evaluated aiming to improve its feasibility for implementation. However, it has predicted that silicon solar cells will continue making an important contribution to the market depending on the maturity of the technology, its availability and especially its cost. For such reasons, the assessment of new alternatives for its production at competitive cost constitutes an opportunity area for research in solar technology (Zweibel et al, 2008; Morales-Acevedo and Casados-Cruz, 2013).

Silicon cells are made from polysilicon materials that can be found as polycrystalline, monocrystalline and amorphous silicon, the former being the most widely used. However, it is important to note that the high cost of polycrystalline silicon is due to both of the processes used to obtain it and the raw material production, silane (SiH_4) (Pizzini, et al., 2005).

One of the processes developed long time ago but currently working for silane production involves the disproportionation of trichlorosilane (obtained from the reaction between metallurgical grade silicon and hydrogen chloride). The metallurgical silicon is the result of natural quartz reduction to silicon tetrachloride and silane (Bakay, 1976). It has been reported that about 40% of the energy required to produce a solar panel it is consumed in the precursor production. Therefore, reduction of energy consumption during the silane production is crucial to minimize the return of investment and thus the cost of the technology.

The conventional process for the silane, dichlorosilane and monochlorosilane production to redistribute trichlorosilane consists of two reactors and multiple distillation columns (Coleman, 1982). The first reactor is used for carrying out the first reaction, where a redistribution of trichlorosilane to dichlorosilane is involved and the second reactor combine components in order to start the redistribution reactions. In addition, four distillation columns are used to separate the products from the reactants, which are then recycled back to the two reactors.

One of the most promising examples of process intensification, is the reactive-separation process which combines reaction and separation in one unit. Harmsen (2007), is a pioneer of the intensification of industrial processes. The author states that the application of reactive distillation technology can save up to 80% of final energy costs associated with capital of reactive and separation sequences.

In this paper several control strategies applied to a conceptual design of a single column of RD for the production of silane, dichlorosilane and monochlorosilane are proposed. Those products are obtained at high purity with commercial and industrial interest using process intensification strategies, assuming the main production target: recovery of the interest components.

Basically the idea of reactive distillation column is improve the chemical conversion, moreover only products are withdrawn from the reactive zone while reactants remain inside the reactive zone for further reaction. Also all material recycles can be eluded and consequently both energy and equipment costs are diminished. Additionally since several freedom degrees are found in a reactive distillation column, such as reflux ratio, total stages, reactive stages and so on, is highly possible to find a single reactive column which might produces all the components produced in the silane redistribution only varying all those freedom degrees.

2. Model of multi-tasking reactive distillation column

The reaction is developed in three steps. In the first one, trichlorosilane ($SiHCl_3$) becomes dichlorosilane (SiH_2Cl_2) and silicon tetrachloride ($SiCl_4$). Subsequently, dichlorosilane reacts to monochlorosilane (SiH_3Cl) and trichlorosilane. Finally, monochlorosilane is converted to silane (SiH_4) and dichlorosilane. The three reaction steps are shown in Equations 1.1 to 1.3.

$$2SiHCl_3 \rightarrow SiH_2Cl_2 + SiCl_4 \quad (1.1)$$

$$2SiH_2Cl_2 \rightarrow SiH_3Cl + SiHCl_3 \quad (1.2)$$

$$2SiH_3Cl \rightarrow SiH_4 + SiH_2Cl_2 \quad (1.3)$$

The kinetic parameters for the involved reactions are taken from Huang et al., (2013). Further, since this is a preliminary study we considered the equilibrium-based RadFrac good enough to describe the process in the Aspen Plus process simulator V.8.4. Also, this equilibrium model would let us know the dynamic behavior that rate-based model would not due to Aspen dynamics are not able to work under those rate-based conditions. For simplicity, the reactions were specified as pseudo-homogenous, occurring in liquid phase.

The design task was performed in two stages. In the first stage, a preliminary design was conducted for defining the operating conditions that allow the reaction selectivity to be displaced in order to obtain the specified purity of each product. These preliminary designs were obtained by performing a parametric sensitivity analysis, varying: reflux ratio, number of total stages, number of reactive stages, feed stage, reboiler duty, pressure, and distillate to feed ratio. Then, the system with the largest internal flows (and consequently with the higher dimensions) was utilized for initializing a second analysis, where the target is to find the necessary conditions for producing all the three components within the same multitasking column.

The obtained results suggested a RD multitasking column with 65 stages, wherein the reaction zone is located between the stages 21 to 50 and the reactant is fed on the plate 51. A feed stream containing $SiHCl_3$ pure with a total feed flowrate of 10 kmol/h at 5.5 atm and 323.15 K, was considered. The column pressure was ranged such that the temperature in the reaction section does not exceed 100°C to avoid the catalyst deactivation and the pressure drop across the column was defined as 0.5 kPa. The holdup volume for the reaction stages was defined as 0.15 cubic meter in order to provide 1.5 min residence time (See Figure. 1).

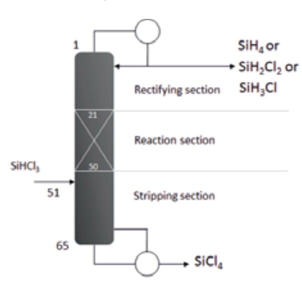

Figure. 1. Multitasking column for the production of silane, dichlorosilane and monochlorosilane.

3. Results

The operating conditions such as pressure, reboiler duty and reflux ratio for each of the components at a molar purity of 0.995 are shown in Table 1, those values marked with a dash [-] represent infeasible operation points. The data in Table 1, shows the same column (Figure 1) operated in a cycle of three different operative conditions varying reflux ratio, column pressure and reboiler heat duty. Those variable values were obtained by varying the pressure of the column top for each component, by setting the purity of the component of interest with the "design spec" tool.

Table 1. Operating conditions of the RD column to purify silane, dichlorosilane and monochlorosilane

	Silane		Monochlorosilane		Dichlorosilane	
Pressure [atm]	Reflux	Reboiler duty [kW]	Reflux	Reboiler duty [kW]	Reflux	Reboiler duty [kW]
0.50	-	-	-	-	17.89	633.3
1.70	106.81	987.2	42.89	815.5	36.72	1196.6
1.80	93.53	866.5	43.26	820.0	38.46	1247.3
1.90	83.68	777.1	43.72	826.3	40.23	1298.7
2.00	76.58	712.5	44.26	834.0	42.04	1350.8
2.10	71.40	665.3	44.86	842.7	43.88	1403.7
2.20	67.52	629.9	45.52	852.5	45.76	1457.5
2.30	64.56	602.7	46.23	863.2	47.67	1512.2
2.40	62.24	581.2	46.98	874.5	49.63	1568.0
2.50	60.39	564.0	47.78	886.6	51.64	1624.8
2.60	58.89	549.9	48.61	899.4	53.69	1682.9
4.50	52.23	477.5	-	-	-	-

As noted, for this reactive distillation there is an operative window wherein the three components can be produced under a range of pressure between 1.7 and 2.6 atm. In order to facilitate the column operation in multitask mode, the pressure column must be adjusted in the same value for all products. This selection cannot be made based only on the energy requirements but also on their dynamic characteristics.

The closed loop policy was performed in accordance with the LV configuration: the control variable is the purity of each component and the manipulated variable is the reflux ratio. The PI controllers are tuned using the tuning criterion of IAE minimization. The obtained results suggest that the best value for the column pressure is 2.3 atm, such that the remaining degrees of freedom for process control are the reboiler duty and reflux ratio.

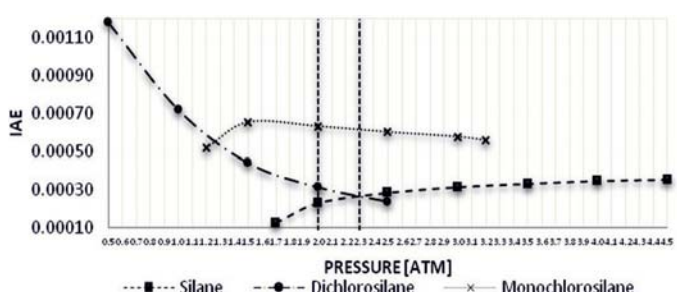

Figure 2. Response of pressure vs value of IAE for each component.

In order to evaluate the dynamic behavior of the multitasking reactive distillation column, three control strategies were implemented: temperature, composition and cascade (temperature/composition), the all structures are reported elsewhere (Luyben, 2008; Gorak, and Hartmut, 2014). The temperature controller was located according to the sensitivity criterion (Luyben, 2006). The most sensitive plates are 3 and 54 for the silane, 9 and 30 for dichlorosilane, and 5 and 41 to monochlorosilane. Figure 3 outlines the cascading arrangement.
The main control objective is to maintain the specified product purities at 99.5 mol%. The disturbances considered are changes in feeding rates and composition.
Figure 4 shows the closed loop responses of all control systems to positive disturbances, as can be observed all three strategies are able to stabilize the composition of the three components, showing good disturbance rejection with similar dynamic behaviour. In some cases the temperature control drives the product to a higher purity value.

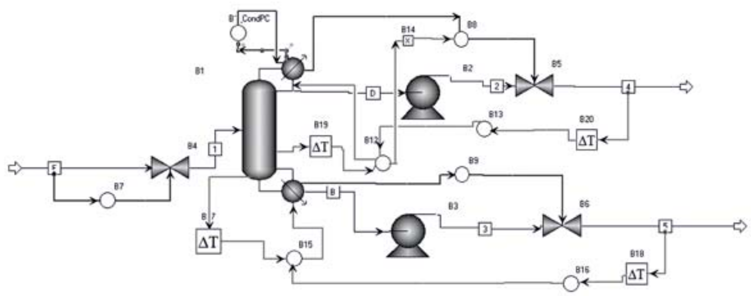

Figure 3. Structure of cascade control.

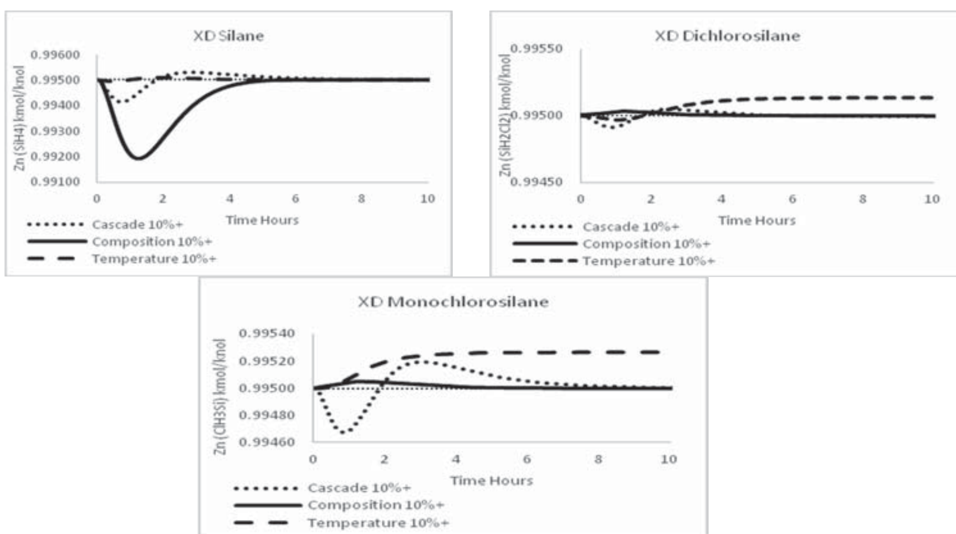

Figure 4. Disturbance in the feed rate (+ 10% F) for a) silane, b) dichlorosilane, c) monochlorosilane [cascade, composition and temperature].

When temperature control strategy is considered, the best dynamic behavior was observed in silane, showing both lower settling time and lower overshoot. However,

other two compounds showed better dynamic properties when a composition control strategy was considered. In general all control strategies showed a good dynamic behavior.
The final selection depends on the control engineer criteria, which needs to find a balance between the optimum dynamic performance and the complexity of the system. In these cases the cascade control strategy represent the most complex structure because of several control devices are required, while the temperature control provides a simple strategy with several advantages for its practical implementation

Conclusions

In this study is showed the feasibility to produce three components (silane, dichlorosilane and monochlorosilane) using a single multitasking reactive distillation column, taking trichlorosilane as raw material. All components were obtained at high purity, having an entire conversion of the trichlorosilane to silane and so on. This process showed an economical advantage in comparison with the conventional process where production and purification are separated operations. In this proposed design, it was possible to switch from a product to other changing only the design variables such as column pressure, reboiler heat duty and reflux ratio. After several tests the column pressure was fixed at 2.3 atm since it was observed the best transient response for all products at this condition. Moreover, three control strategies were evaluated, the obtained results suggest that all of them are able to stabilize the process with similar dynamic performance.

References

A. Gorak, S. Hartmut, 2014, “Distillation: Operation and Applications”, Elsevier, Oxford, UK, pp 1-37.

A. Morales-Acevedo, G. Casados-Cruz, 2013, “Forecasting the development of different solar cell technologies”, Int. J. Photoenergy., pp. 1-5.

A.F.B. Braga, S. P. Moreira, P.R. Zampieri, J.M.G. Bacchin and P.R. Mei, 2008, “New Processes for the Production of Solar-Grade Polycrystalline Silicon: A Review”, Sol. Energy Mater. Sol. Cells, 92, 418.

C. J Bakay, 1976, “Process for Making Silane”. U.S. Patent 3,968,199.

G. J., Harmsen, 2007, Reactive Distillation: The Front-Runner of Industrial Process Intensification: A Full Review of Commercial Applications, Research, Scale-up, Design and Operation. Chem. Eng. Process, 46, 774−780.

H. Xun, D. Wei-Jie , Y. Jian-Min, and X. Wen-De, 2013, “Reactive Distillation Column for Disproportionation of Trichlorosilane to Silane: Reducing Refrigeration Load with Intermediate Condensers,” Ind. Eng. Chem. Res., 52, pp. 6211−6220.

K. Zweibel, J. Mason and V. Fthenakis, 2008, “Solar Grand Plan: Solar as a Solution”, Sun&Wind Energy, 4 pp. 112-117.

K.M. , Guthrie, 1969, Capital cost estimating, Chemical engineering, March 24, pp. 114-142.

L. Coleman,, 1982, Process for the production of ultrahigh purity silane with recycle from separation columns. (Union Carbide Corporation) US 4,340,574. DE 33 11 650.

P.Woditsch, W. Koch, 2002, “Solar grade silicon feedstock supply for PV industry,” Solar Energy Materials & Solar Cells, 72, pp. 11–26.

S. Pizzini, M. Acciarri, S. Binetti, 2005, “From electronic grade to solar grade silicon: chances and challenges in photovoltaics”, Phys. Status Solidi A, 202, pp. 2928-2942.

W.L. Luyben, C.C. Yu, “Reactive Distillation Design and Control”, Wiley-AIChE (2008).

W.L.Luyben, “Evaluation of criteria for selecting temperature control trays in distillation columns”, Journal of Process Control, 16 (2) (2006), pp. 115–134.