



## Optimization of a reactive distillation process with intermediate condensers for silane production

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### ABSTRACT

This work presents a reactive distillation column for the catalytic disproportionation of trichlorosilane to silane which includes three consecutive reversible reactions. This reaction system is however characterized by a large distinction in the boiling points of the components, which make the reactive distillation extremely favored. Nevertheless, the normal reactive distillation column possesses the shortage of high refrigeration requirement. By removing heat at temperature higher than that at the condenser a superstructure representation, rigorous simulations, and optimization problems were combined to derive optimal reactive distillation columns which can realize heat integration between stages and utilities at several refrigeration conditions. An iterative simulation-optimization procedure was proposed to consider temperature changes in stages due to heat integration. The results showed that the installation of two inter-condensers results in the best option with economic savings up to 56%.

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### 1. Introduction

Global power consumption currently stands at approximately 15 TW, the vast majority of which is generated by the combustion of fossil fuels. The associated release of CO<sub>2</sub> from these anthropogenic sources has dramatically altered the composition of the atmosphere and may detrimentally impact global temperature, sea levels, and weather patterns (Parida et al., 2011). Furthermore, the realization that fossil fuels are not inexhaustible and that enhancing recovery of coal, oil and natural gas presents additional risks, drive energy policy scenarios that are based on renewable forms of energy. In most of the proposed scenarios, solar energy is the primary constituent as it is the major energy source over large regions. Solar energy in addition to maintaining life on the planet, is used on demand in three basic forms based on anthropogenic processes: Electricity from the direct conversion of solar energy using semiconductor materials (solar photovoltaics,

PV); electricity from captured thermal energy (concentrated solar power), and heat from the sun (solar thermal). Overall, the market for photovoltaics has been growing at an average rate of 45% per year over the past decade (Braga et al., 2008; Huang et al., 2013).

The five year growth rate from 2007 to 2011 was approximately 70% per year, but this was slowed down to 15% in 2012 as the incentives in several European countries were reduced. While the growth numbers are very impressive, the 27 GW installed in 2011 is just a fraction of one percent of the total amount of electricity that was generated by all sources, indicating that there is plenty of room for further growth (Müller et al., 2006). Photovoltaics use semiconductor materials to generate electricity from solar energy. A semiconductor is a solid, mostly crystalline, material such as silicon, selenium or germanium. Semiconductors have electrical conductivities greater than insulators but lower than metals which are good conductors. At low temperatures and high level of purity, they are insulators but at high temperatures and/or when they are doped and excited by sunlight, they conduct electrons. The most commonly used semiconductor element is silicon. Therefore silicon, the starting material for the crystalline wafer, today is the most important material in PV industry (Fthenakis et al., 2009).

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To widely commercialize the solar technology, one challenge faced by the PV industry is to decrease the manufacturing costs, especially the cost of solar-grade silicon feedstock material. The most well-known chemical route to produce solar-grade silicon is the Siemens process, which performs decomposition at about 1000 °C of trichlorosilane by Chemical Vapor Deposition. However, the extremely high consumption of electrical energy as well as the corrosion caused by the byproduct (hydrochloric acid) in some way has limitations in the production of low-cost polysilicon.

The silane pyrolysis process operated in a continuous fluidized bed reactor for its low consumption of energy is an alternative (Iya, 1989; Flagella, 1992). In order to reduce the temperature of silicon deposition, silane (SiH<sub>4</sub>) is used as starting source in the CVD reactor. Other very well known route to produce silane is the disproportionation reactions of SiHCl<sub>3</sub> involving the formation of dichlorosilane (SiH<sub>2</sub>Cl<sub>2</sub>) and monochlorosilane (SiH<sub>3</sub>Cl) as intermediates and tetrachlorosilane (SiCl<sub>4</sub>) as by product (Bakay, 1976). The conventional process for highly pure silane production from trichlorosilane disproportionation is through two reactors and several separation units. However, due to unfavorable chemical equilibrium, this reaction and separation process requires an extremely large recycle ratio and thus the cost of energy as well as investment is very high (Breneman, 1987).

In this context, Reactive Distillation (RD) is particularly attractive for reactions limited by chemical equilibrium, and it can be suitable for the disproportionation reactions because it eliminates conversion and phase equilibrium limitations by continuous removal of products from the reaction zone unit (Sundmacher and Kienle, 2003).

Reactive distillation is a combination of separation and reaction in a single vessel. The concept of combining these two important functions for enhancement of overall performance is an important concept in chemical engineering. The commercial success of reactive distillation for the production of MTBE was immediately followed by another remarkable achievement with the Eastman Kodak process that condensed the whole chemical plant for methyl acetate in a single reactive distillation unit (Sundmacher and Kienle, 2003).

In some applications, particularly in cases where thermodynamic reaction equilibrium prevents high conversions, the coupling of distillation to remove the reaction products from the reaction zone can significantly improve the overall conversion and selectivity at pressures and temperatures that are compatible with the distillation conditions. In other applications, the presence of reactions is used to overcome the separation problems caused by the presence of azeotropes. Therefore, this combination of reaction and distillation often results into simpler and intensified processes with less recycle streams, reduced waste handling and, as a consequence, lower investment and operating costs (Carrera-Rodríguez et al., 2011).

Some important references related to reactive distillation and silanes production can be described. Bakay (1976) proposed in a patent, the process for manufacturing silane in a bed of anion exchange resin by controlling the top temperature between the boiling points of silane and trichlorosilane. In such process, silane could be produced directly from trichlorosilane in a single step process wherein a solid redistribution catalyst also served as the contact surface in a fractional distillation column. The Bakay's patent, although it was not so named as Reactive Distillation, the process was "the essential embodiment because that both chemical reaction and distillation separation were conducted in the same apparatus" (Breneman, 2013).

Yamada et al. (1984) described a process using amines as homogeneous catalysts. Inoue (1988) reported in a patent a Reactive distillation process for dichlorosilane redistribution to silane, trichlorosilane and tetrachlorosilane. Matthes et al. (1988)

**Table 1**  
Binary interaction parameters (Huang et al., 2013).

<i>i</i>	<i>j</i>	<i>k<sub>ij</sub></i>
SiCl <sub>4</sub>	SiHCl <sub>3</sub>	0.01603
SiCl <sub>4</sub>	SiH <sub>2</sub> Cl <sub>2</sub>	0.02108
SiHCl <sub>3</sub>	SiH <sub>2</sub> Cl <sub>2</sub>	0.05183
SiH <sub>2</sub> Cl <sub>2</sub>	SiH <sub>3</sub> Cl	-0.00538
SiH <sub>3</sub> Cl	SiH <sub>4</sub>	0.000953

and Frings (2000) reported a distillation process with a coupled reactor for trichlorosilane redistribution to dichlorosilane and tetrachlorosilane.

Later, Müller et al. (2002) studied potential reactive distillation schemes for silane production without paying attention to the generation of intermediate reaction products.

Muller et al. (2005) and Block et al. (2005) with Solarworld AG suggested some reactive distillation schemes with inter-condensers. Sonnenschein et al. (2011) with Envonik Degussa presented a distillation column with a side reactor.

It can be considered that one of the major disadvantages in using reactive distillation for the disproportionation of trichlorosilane is the low boiling point of the target product at the top of the column (silane), which is around -112 °C and enforces the use of refrigeration services to condensate it. To alleviate this issue, recently, the use of intermediate condensers has been proposed to reduce the energy consumption and cost of the refrigeration service at the top of the column (Muller et al., 2005; Huang et al., 2013).

On the other hand, Huang et al. (2013) have reported a design procedure to derive feasible reactive distillation columns for silane production from trichlorosilane by means of sensitivity analysis, a trial-and-error method, and some arbitrary assumptions. In addition, Breneman (2013) proposed the used of reactive distillation for the production of several hydrohalosilanes, however, the design analysis in terms of energy consumption was not considered, therefore there is a good area for energy and cost reduction in the aforementioned reactive distillation schemes.

In this work, we investigate the feasibility of applying reactive distillation to the production of silane through the disproportionation of trichlorosilane. This reaction system is however characterized by a large distinction in the boiling points of the components, which makes the reactive distillation extremely favored. Nevertheless, the normal reactive distillation column possesses the shortage of high refrigeration requirement due to the standard boiling point of the overhead product. The design and optimization of reactive distillation column with inter-condensers has been studied as a feasible alternative to alleviate this drawback.

## 2. Case study

The production of silane (SiH<sub>4</sub>) through disproportionation of trichlorosilane (SiHCl<sub>3</sub>) in a reactive distillation column has been studied. The reaction occurs in three steps, having as intermediates dichlorosilane (SiH<sub>2</sub>Cl<sub>2</sub>) and monochlorosilane (SiH<sub>3</sub>Cl) and as by-product chlorosilane (SiCl<sub>4</sub>), as follows:



The vapor-liquid equilibrium has been modeled using the Peng-Robinson equation of state, with the binary interaction parameters shown in Table 1.

The reaction occurs in liquid phase, using as catalyst the polymeric resin Amberlyst A-21. For the disproportionation of

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