

Modelling and Measuring Silane Gaseous Growth Pathways in Pilot-Scale Silicon Production Reactors

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Industrial reactors around the world use SiH_4 and SiHCl_3 feedstocks to produce high-grade silicon that ends up in our computers, solar cells, and even the batteries in our electric cars. We seek to improve our understanding, and thus better control, the conversion of gaseous SiH_4 to solid Si by combining chemical kinetics models, reactor models and measurements of the silicon-production reactors in our laboratory.

Gas-phase conversion from silane (SiH_4) to macroscopic solid silicon involves many Si-Si chain growth steps to make polysilane structures, and subsequent H_2 elimination leads to thermodynamically stable solid silicon. There are many possible routes leading to macroscopic silicon. Here we focus on the gas-phase conversion process, whose chemical formula $\text{SiH}_4(\text{g}) \rightarrow \text{Si}(\text{s}) + 2\text{H}_2(\text{g})$ hides much of the overall complexity. Important mechanisms in the conversion from SiH_4 to solid Si include H_2 elimination, Si-Si chain propagation by silyl (SiH_2) insertion to Si-H bonds, 1,2-H shifts in unsaturated silyl species, and silane ring formation. The number of possible intermediates and pathways explodes quickly as the silane chains grow.

Our approach to tackling this problem is three-fold. First, we generate a reaction map, estimate Arrhenius parameters for each reaction step using group additivity schemes available in the literature, and propagate the concentrations of intermediates using procedurally generated rate equations from our model. Second, we couple the chemical conversion simulation with physical models of the reactors using our own software. Third, we measure the polysilanes produced in continuous-flow silane reactors using gas chromatography-mass spectrometry measurements to serve as feedback to, and verification of, the modelling. We vary the temperature, pressure, and reactant gas mixtures of the lab-scale reactors to examine their effects on the chemistry. As we explore this broad parameter space with the model, we gain competence which can be used to optimize not only our experiments but, in the end, industrial silicon production reactors.