SURFACE AND GAS PHASE CHEMISTRY OF THE MOCVD OF ZnSe

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ABSTRACT

Quantum chemistry was used to investigate gas phase and surface chemistry active during the MOCVD of ZnSe from H_2Se and $Zn(CH_3)_2$ (1-2). Energies, structures and transition state geometries were determined with density functional theory. It was found that the direct reaction between the reactants is too slow to explain the formation of adducts experimentally observed (1).

Then an alternative pathway, based on a radical chain mechanism started from the desorption of CH_3 radicals from the surface was proposed. The kinetic constants for this pathway were calculated and found to have almost no activation energy. Simulation of an experimental reactor (table I) showed that this mechanism is able to explain the formation of adducts in the gas phase experimentally observed (Fig 1-2). In fig. 3a mole fraction of $H(SeZn)_3CH_3$ as a function of chamber pressure is showed. It can be observed that adducts are produced both at low and high pressure. It was found that the formation of adducts is proportional to the inlet reactants mole fraction (represented in fig. 3b). Finally the mole fraction of $H(SeZn)_3CH_3$ produced as a function of temperature and total flow rate was examined. The results reported in fig. 3c show that, as expected, to an increase of temperature corresponds an increase of the adduct formation rate. In fig. 3d the dependence of the adducts mole fraction from the total flow rate is sketched.

$N(Et)_3$ is usually added to $Zn(CH_3)_2$ to diminish the extent of pre-reactions in the gas phase (3). The effect that $N(Et)_3$ has on gas phase reactivity was previously investigated (1). It was found that the reason why $N(Et)_3$ diminishes the formation of adducts is that it strongly interacts with ZnSe molecules as soon as methyl groups are replaced by Se groups, thus preventing, through steric repulsion, their progressive growth (1). The kinetic scheme previously developed was linked with the reactions of $N(Et)_3$ with ZnSe adducts; kinetic constants were calculated by collisions theory. The kinetic scheme was finally embedded in the reactor above described and results of the simulation showed that ZnSe adducts are practically absent in the reaction chamber when $N(Et)_3$ is introduced .

REFERENCES

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Table I. Reactor geometry and operating conditions adopted to perform fluid dynamic simulation.

Susceptor Length	12 cm
Reactor Chamber height	3.5 cm
Reactor Chamber width	9 cm
Operative pressure	200 torr
Total flow rate	2000 sccm
Carrier gas	Hydrogen
$(CH_3)_2Zn$ inlet mole fraction	10^{-3}
H_2Se inlet mole fraction	10^{-3}
Susceptor temperature	260° C

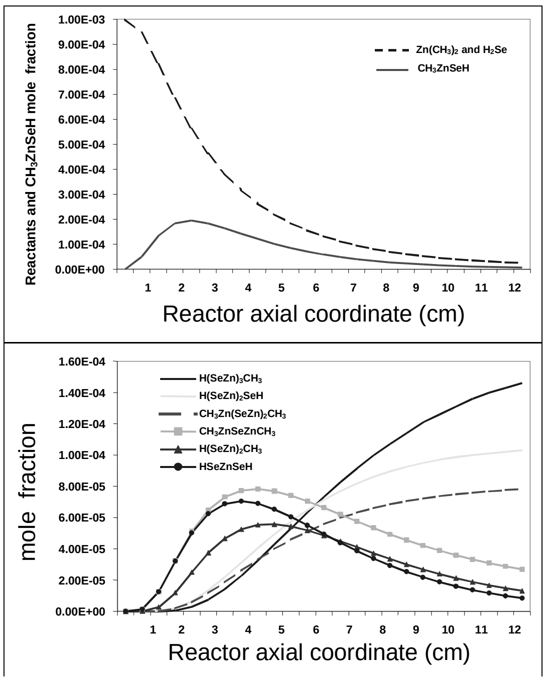

Fig. 1-2. Comparison between the mole fraction of reactants and CH_3ZnSeH produced in the gas adopting the radical chain mechanism and the mole fraction of adducts produced. Simulation performed with the conditions specified in table I.

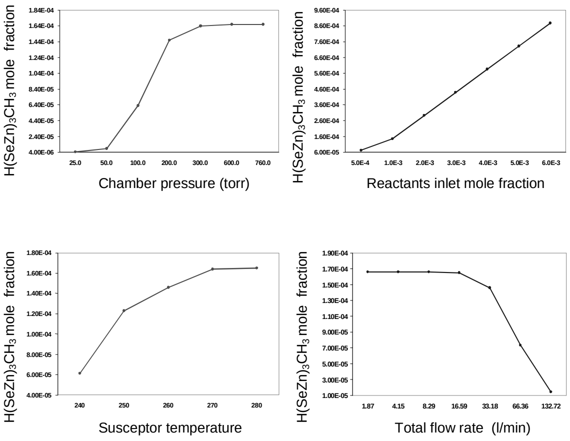

Fig.3a. Calculated mole fraction of $H(SeZn)_3CH_3$ produced as a function of chamber pressure; 3b inlet mole fraction; 3c. susceptor temperature; 3d. total flow rate.