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References

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Investigations into disilane; rate constants, isomers, and line-lists.

Reaction Mechanisms | Orientational effects | Molecular Dynamics | DFT-B | Monte-Carlo type statistics | •••

Abstract

2 hrs • ★

The reaction $SiH_2+SiH_4 \rightarrow Si_2H_6$ is of importance to industry, especially plasma etching. Investigations into the reaction, including rate constants calculated using a statistical Monte-Carlo type density functional tight-binding (DFT-B) approach using deMonNano [1], are discussed here. An interesting result was the formation of multiple conformers of disilane, separated by an energy barrier of 20.37 kJ/mol. Current work also includes the calculation of a molecular line-list for SiH_2.

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Methodology

3 hrs • ★

Over 100,000 DFT-B molecular dynamics simulations were performed with deMonNano by firing SiH_2 at SiH_4 and recording if the simulation resulted in the formation of Si_2H_6. To allow full sampling of possible collision configurations, a z-matrix was used to set the initial geometries. The schematic, left, shows how the molecules are randomised, with the impact parameter, *b,* two angles, α, four dihedral angles, γ, and the velocity of the SiH_2, ν, randomised for each collision simulation, totalling 9 variable parameters inc. temperature.

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Results ► Discussion

6 hrs • ★

The rate constants calculated with the equation shown on the left are shown below, where they are compared to experimentally determined rate constants from literature [2]. The values are correct to the same order of magnitude for both temperatures, which is an excellent result for this level of theory.

Comparison of literature and calculated rate constants for $SiH_2+SiH_4 \rightarrow Si_2H_6$ [2]

300 K		600 K	
DFT-B	Experimental	DFT-B	Experimental
1.06×10^{-9} $cm^3\ s^{-1}$	2.13×10^{-10} $cm^3\ s^{-1}$	8.61×10^{-10} $cm^3\ s^{-1}$	5.40×10^{-11} $cm^3\ s^{-1}$

The barrier height of isomerisation was calculated with MOLPRO [3] and was found to be 20.37 kJ/mol (7,123.78 cm^{-1}) more than the umbrella geometry, and 249.64 kJ/mol (20,867.93 cm^{-1}) more than the bowtie structure [see figure on left for definition of structures]

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Conclusion

8 hrs • ★

The rate constants calculated with deMonNano at 300 K and 600 K are in good agreement with the experimental literature values, and can be calculated quickly for the $SiH_2+SiH_4 \rightarrow Si_2H_6$ system at pressures of 0.1 MPa. The calculated and experimental rate constants show the same trend with temperature, hence it would be possible to extrapolate rate constant values to temperatures which have not been experimentally determined. The Umbrella→Bowtie barrier height between the two isomers was found to be 20.37 kJ/mol.

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