

Contents

Abbreviations	v
List of Symbols	vii
Abstract	ix
Kurzfassung	xi
1. Introduction	1
2. Predictions in Homogeneous Liquid Phases and Neutral Solutes	6
2.1. Introduction	6
2.2. Methodology	8
2.2.1. Reaction Details	8
2.2.2. Computational Details	10
2.2.3. Calculation of the NIPAM Reaction Rate Constants	13
2.3. Results and Discussion	15
2.3.1. Validation of the Computational Results	15
2.3.2. Reaction Rate Constants	16
2.3.3. Enthalpy of Reaction	21
2.4. Conclusion	23

3. Predictions in Homogeneous Liquid Phases with Charged Solutes: The Bottleneck in Predicting Accurate Reaction Rate Constants in the Liquid Phase When Ions are Involved	25
3.1. Introduction	26
3.2. Methodology	30
3.2.1. Accounting for Uncertainty in $\Delta G_{\text{solv}}(\text{H}^+)$	30
3.2.2. COSMO-RS	32
3.2.3. Cluster Continuum Model	34
3.2.4. COSMO-RS-ES	35
3.3. Results and Discussion	36
3.3.1. COSMO-RS	42
3.3.2. Cluster Continuum Model	43
3.3.3. COSMO-RS-ES	45
3.4. Conclusions	46
4. Predictions in Inhomogeneous Liquid Phases Based on Reactive Molecular Dynamics Simulations: Statistical Uncertainty and Pa- rameterization	49
4.1. Statistical Uncertainty of Rate Constants from Reactive MD Simulations	51
4.1.1. Reaction Events	53
4.1.2. Applications	56
4.1.3. Statistical <i>vs.</i> methodological errors	63
4.1.4. Conclusions about the Statistical Uncertainties of Reaction Events	65
4.2. Force Field Applicability	65
4.2.1. Methodology	67
4.2.2. Results and Discussion	68
4.2.3. Conclusions About Force Field Applicability	71
5. Conclusions and Outlook	73
Bibliography	78

A. Appendix	100
A.1. Supporting information to Chapter 2	100
A.1.1. Additional Figures	103
A.1.2. Mayo-Lewis-Plots	108
A.2. Supporting Information to Chapter 3	109
A.2.1. Calculated Data	109
A.3. Supporting Information to Chapter 4	135
A.3.1. Confidence Interval - Detailed Derivation	135
B. CV and List of Journal Publications	136