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DEVELOPMENT AND IMPEMENTATION OF QUANTUM-CLASSICAL STOCHASTIC METHODS FOR THE CALCULATION OF KINETIC RATES OF ELEMENTARY REACTIONS IN SIMPLE AND COMPLEX CONDENSED PHASE

This work develops a model for interpreting macroscopic kinetic constants in elementary reactions, particularly when mass transport and chemical reactions occur on comparable timescales, as in enzymatic catalysis. We focus on bimolecular exchange reactions, specifically SN2 nucleophilic substitutions in haloalkanes, using bromide and chloromethane as a case study. Our approach employs a reaction-diffusion model inspired by Sevein and Moro [1], integrating mass transport with localized quantum-mechanical reactions. The governing equations consist of (i) a diffusive term, dependent on the diffusion coefficient and mean field interaction potential determined via steered molecular dynamics [2], and well-described by a modified Buckingham potential and (ii) a reactive term, modeled through a master equation parameterized with microscopic kinetic constants [3,4].

This model establishes a continuous description of macroscopic kinetic constants by linking the interplay of reaction and transport timescales. Despite simplifications such as neglecting rotational dynamics and separating barrier crossing from other relaxation processes the method accurately captures expected kinetic behaviors across a wide dynamic range. It estimates macroscopic kinetic constants in an ab initio fashion by correlating key molecular parameters: diffusion coefficients, interaction potentials, and microscopic rate constants. This framework enhances our understanding of reaction mechanisms in complex environments and provides a basis for future studies on reactive dynamics in liquids.

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