

Ab Initio Molecular Dynamics Simulations of Phosphate Hydrolysis Using Neural Network Potentials

Albert MAKHMUDOV

Supervisor: Prof. J. Harvey
KU Leuven

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Foreword

Contribution statement

Summary

List of abbreviations

Contents

1	Introduction	1
1.1	Role of phosphates in biological systems	1
1.2	Enzymes involved in phosphate hydrolysis	1
1.3	Reaction mechanism	1
1.4	Research goals	1
2	Theory	2
2.1	A brief introduction to statistical mechanics	3
2.1.1	Classical forcefields and molecular dynamics	3
2.1.2	The canonical ensemble and free energy calculations	3
2.1.3	Enhanced sampling techniques	3
2.2	Density functional theory	3
2.2.1	The Kohn-Sham approach	3
2.2.2	Generalised gradient approximation and PBE functional	3
2.2.3	<i>Ab initio</i> molecular dynamics and GPW method	3
2.3	Extended tight binding	3
2.4	Neural network potentials	3
2.4.1	Deep neural networks	3
2.4.2	Invariance and equivariance	3
2.4.3	Behler-Parrinello neural network potentials	3
2.4.4	Equivariant neural network potentials	3
3	Computational Details	4
3.1	Training dataset generation	4
3.1.1	System preparation	4
3.1.2	Initial equilibration using the classical forcefields	4
3.1.3	xTB based exploration of the configuration space	4
3.1.4	Data labeling	4
3.1.5	Iterative training of the neural network potential	4
3.2	Production runs at different temperatures	4

<i>CONTENTS</i>	viii
3.3 Validation of the transition states	4
3.4 Data analysis and visualisation	4
4 Results and Discussion	5
5 Conclusions	6
Bibliography	7
A Supplementary information	8

Chapter 1

Introduction

- 1.1 Role of phosphates in biological systems**
- 1.2 Enzymes involved in phosphate hydrolysis**
- 1.3 Reaction mechanism**
- 1.4 Research goals**

Chapter 2

Theory

2.1 A brief introduction to statistical mechanics

2.1.1 Classical forcefields and molecular dynamics

2.1.2 The canonical ensemble and free energy calculations

2.1.3 Enhanced sampling techniques

Metadynamics and its well-tempered flavour

Kinetics from metadynamics

2.2 Density functional theory

2.2.1 The Kohn-Sham approach

2.2.2 Generalised gradient approximation and PBE functional

2.2.3 *Ab initio* molecular dynamics and GPW method

2.3 Extended tight binding

2.4 Neural network potentials

2.4.1 Deep neural networks

Multilayer perceptron

Graph neural networks

Message passing neural networks

2.4.2 Invariance and equivariance

2.4.3 Robust Perrinello neural network potentials

Chapter 3

Computational Details

3.1 Training dataset generation

3.1.1 System preparation

3.1.2 Initial equilibration using the classical forcefields

3.1.3 xTB based exploration of the configuration space

3.1.4 Data labeling

3.1.5 Iterative training of the neural network potential

First round

Second round

Third round

3.2 Production runs at different temperatures

3.3 Validation of the transition states

3.4 Data analysis and visualisation

Chapter 4

Results and Discussion

Chapter 5

Conclusions

Bibliography

Appendix A

Supplementary information

Quantum Chemistry and Physical Chemistry

Celestijnenlaan 200F bus 2404

3001 LEUVEN, BELGIË

tel. + 32 16 37 21 98

jeremy.harvey@kuleuven.be

www.kuleuven.be

