SLATT UNDERGRADUATE RESEARCH FELLOWSHIP
FINAL REPORT

SCHOLAR NAME:	Audrey Miles
FACULTY ADVISOR:	Professor William Schneider
PROJECT PERIOD:	Winter Session 2021 (12/01/2020 – 02/03/2021)
PROJECT TITLE:	Benchmarking Ammonia Synthesis Entropies in Microkinetic Modeling
CONNECTION TO ONE OR MORE ENERGY-RELATED RESEARCH AREAS (CHECK ALL THAT APPLY):	**(X) Energy Conversion and Efficiency ( ) Sustainable and Secure Nuclear ( ) Smart Storage and Distribution ( ) Transformation Solar ( ) Sustainable Bio/Fossil Fuels ( ) Transformative Wind**

MAJOR GOALS AND ACCOMPLISHMENTS

Summarize your research goals and provide a brief statement of your accomplishments (no more than 1-2 sentences). Indicate whether you were able to accomplish your goals by estimating the percentage completed for each one. Use the next page for your written report.

RESEARCH GOALS	ACTUAL PERFORMANCE AND ACCOMPLISHMENTS	% OF GOAL COMPLETED
Create a Python notebook capable of evaluating the Schrödinger equation for a 1D arbitrary potential, as well as corresponding thermodynamic quantities.	I developed a Python notebook that utilizes finite difference methods (a numerical technique) to evaluate the Schrödinger equation for any arbitrary potential, and used this notebook to analyze various models that are often used to study catalytic reactions on surfaces including the free translator and harmonic oscillator. I also included several functions that calculate the translational partition function, Helmholtz free energy, and entropy of the given system.	90%
Construct a simple CSTR model studying the evolution of reactant and product concentrations over time.	I developed a simulation that a two-reaction system in an isothermal continuous stirred-tank reactor (CSTR) using stochastic sampling and compared its output to that of a system solved using an analytical solution. This simulation can be expanded to be representative of multi-reaction systems.	100%
Evaluate the rates of ammonia synthesis under various entropy conditions and develop a microkinetic model of the overall catalytic process.	I have begun expanding my model of the multi-reaction system to be representative of the elementary reactions that compose the synthesis of ammonia. By modifying this model to include rate constants as supplied by DFT calculations, I will be able to evaluate the production of ammonia under various entropy conditions.	10%

RESEARCH OUTPUT

Please provide any output that may have resulted from your research project. You may leave any and all categories blank or check with your faculty advisor if you are unsure how to respond.

CATEGORY	INFORMATION
EXTERNAL PROPOSALS SUBMITTED	(Sponsor, Project Title, PIs, Submission Date, Proposal Amount) N/A
EXTERNAL AWARDS RECEIVED	(Sponsor, Project Title, PIs, Award Date, Award Amount) N/A
JOURNAL ARTICLES IN PROCESS OR PUBLISHED	(Journal Name, Title, Authors, Submission Date, Publication Date, Volume #, Page #s) N/A
BOOKS AND CHAPTERS RELATED TO YOUR RESEARCH	(Book Title, Chapter Title, Authors, Submission Date, Publication Date, Volume #, Page #s) N/A
PUBLIC PRESENTATIONS YOU MADE ABOUT YOUR RESEARCH	(Event, Presentation Title, Presentation Date, Location) N/A
AWARDS OR RECOGNITIONS YOU RECEIVED FOR YOUR RESEARCH PROJECT	(Purpose, Title, Date Received) N/A
INTERNAL COLLABORATIONS FOSTERED	(Name, Organization, Purpose of Affiliation, and Frequency of Interactions) N/A
EXTERNAL COLLABORATIONS FOSTERED	(Name, Organization, Purpose of Affiliation, and Frequency of Interactions) N/A
WEBSITE(S) FEATURING RESEARCH PROJECT	(URL) N/A

OTHER PRODUCTS AND SERVICES (e.g., media reports, databases, software, models, curricula, instruments, education programs, outreach for ND Energy and other groups)	(Please describe each item in detail) In collaboration with Craig Waitt, I am refining the Python notebook that I developed to evaluate the Schrödinger equation and corresponding thermodynamic quantities to be available as a software package. We intend to eventually publish this software package through the Journal of Chemical Education.

RESEARCH EXPERIENCE

Please let us know what you thought of your research experience: Did this experience meet your expectations? Were lab personnel helpful and responsive to your needs? What else could have been done to improve your experience or achieve additional results?

Through the Slatt Undergraduate Research Fellowship, I was afforded an excellent research experience. Despite the uncertainties and laboratory restrictions caused by the COVID-19 pandemic, I was warmly welcomed by Professor William Schneider's research group. Through my collaboration with doctoral student Craig Waitt, I have been exposed to a number of computing techniques and chemical concepts. My experience in presenting my work to the group and communicating my research has been invaluable, and the guidance that I have received from both Professor Schneider and Craig has truly helped me develop my own abilities and my sense of the overall research community. The Slatt Fellowship allowed me to devote my time to researching remotely, which greatly increased my progress and productivity. The strictly virtual nature of the work certainly presents challenges, but my actual research has been relatively unaffected. I am very grateful to have been able to spend time simply learning about the world around me through the lens of computational and chemical science with the help of outstanding mentors, as made possible by the Slatt Fellowship.

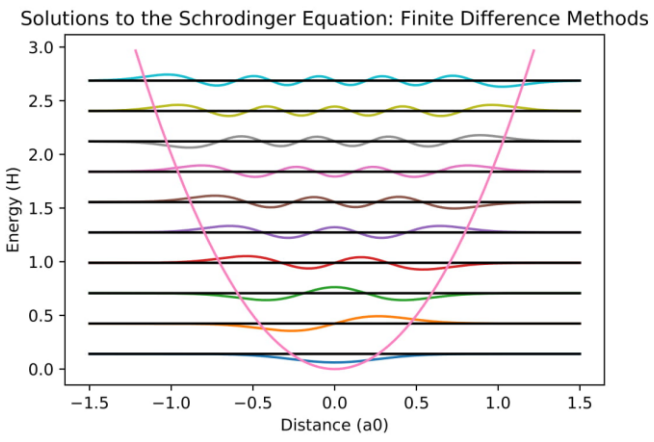
FINAL WRITTEN REPORT

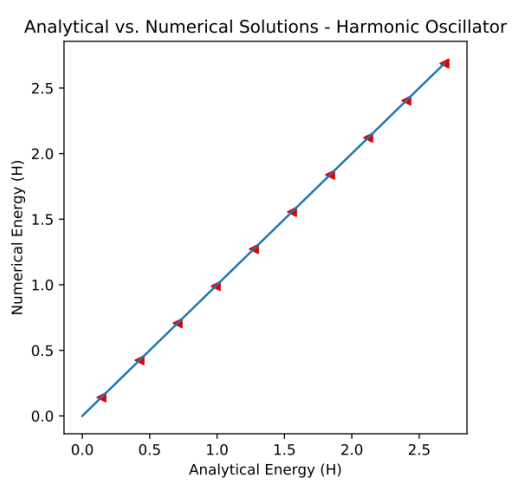
(Please use the space below to describe your research project and objectives, any findings and results you can share, and graphs, charts, and other visuals to help us understand what you achieved as a result of this research experience.)

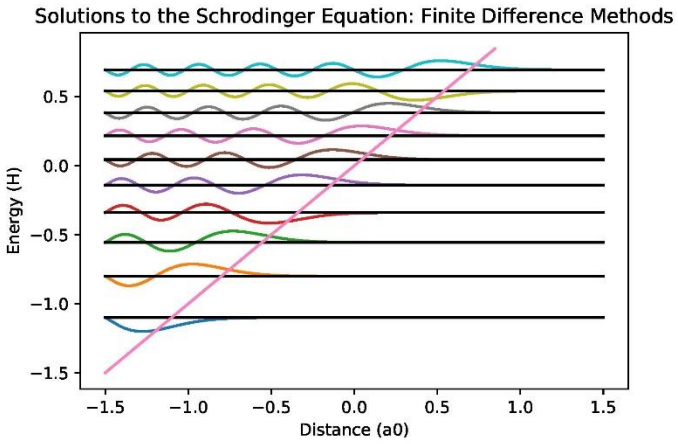
In order to achieve the eventual goal of collaborating with doctoral student Craig Waitt to benchmark the ability of various models to accurately calculate the entropic contributions of ammonia synthesis and develop microkinetic models describing the overall catalytic process, it was necessary to pedagogically develop an understanding of these models.

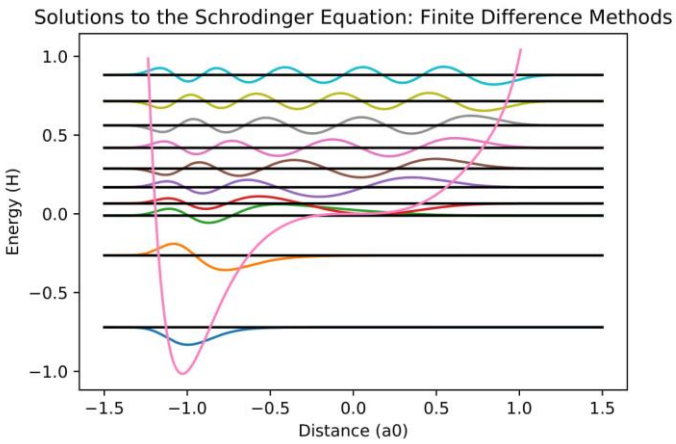
The free energy (and ultimately entropy) calculations under consideration are obtained by combining density functional theory (DFT) energies with standard models describing the properties of systems. These models are defined by the shape of the potential energy function that determines the potential energy at any given point in the system. For example, the harmonic oscillator model is often used to describe a system in which a particle is adsorbed onto a surface and is represented by a parabolic potential. In order to understand these models, a series of Python programs were written that evaluated Schrödinger's wave equation analytically for a variety of potentials. These solutions to the Schrödinger equation take the form of wavefunctions that describe all wave characteristics of the system and corresponding energies, which then allow for the calculation of thermodynamic quantities like entropy.

However, as the potential energy function becomes more complex, it becomes increasingly difficult to solve the Schrödinger equation analytically and numerical methods are necessary. Finite difference methods, a class of numerical techniques that aim to solve differential equations using differential quotients, are effective in solving the Schrödinger equation. By discretizing the domain, a series of linear equations can be generated and solved using matrix algebra techniques. Using these techniques, a Python notebook was written that evaluates the Schrödinger equation for any potential. This code was then applied to various standard models, including the harmonic oscillator, the ideal gas, and several miscellaneous potentials. By comparing my previous analytical results with the numerical results for the particle in a box and the harmonic oscillator, I was able to confirm the ability of the numerical methods to solve the Schrödinger equation.



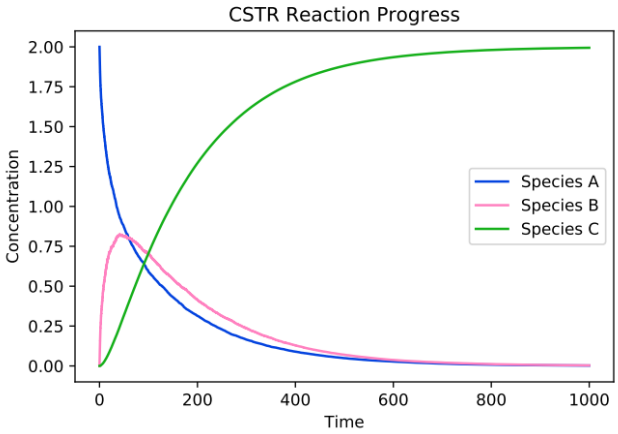






Once the solutions to the Schrödinger equation have been obtained, it is possible to utilize those energy values to calculate thermodynamic quantities including the translational partition function, the Helmholtz free energy, and the entropy of the system. Several functions were appended to the Python notebook that evaluated the Schrödinger equation to calculate these values. Due to its pedagogical nature, I am currently working with Craig Waitt to refine this Python notebook to be distributed as a software package. We are also potentially looking to publish this work in the American Chemical Society's Journal of Chemical of Education.

Following the completion of this Python notebook, it was necessary to develop a foundational understanding of the progress of a reaction in a continuous stirred-tank reactor. In order to pursue future work related to catalytic ammonia synthesis, an understanding of the design and operation of the reactor in which this process occurs is crucial. For this reason, a Python notebook was created that analytically calculates the concentrations of species in a single-reaction system as it occurs in an isothermal CSTR. In order to generalize this model to chemical systems composed of more than one reaction, a second Python notebook was written to numerically solve the increasingly-complex differential equations expressing the concentrations of species in a CSTR over time. This code successfully modeled a two-reaction reaction system in a CSTR.



This CSTR model will form the basis for a microkinetic model describing the process of catalytic ammonia synthesis. The model will be expanded from a two-reaction system to the number of elementary reactions composing the process under consideration, and rate constants will be derived from DFT calculations (as provided by Craig Waitt). This model has already been extended from two to three reactions, and I am working to develop the elementary reactions to be used. This is the model that will soon be used to benchmark the ability of various models to accurately calculate entropic contributions to ammonia synthesis.