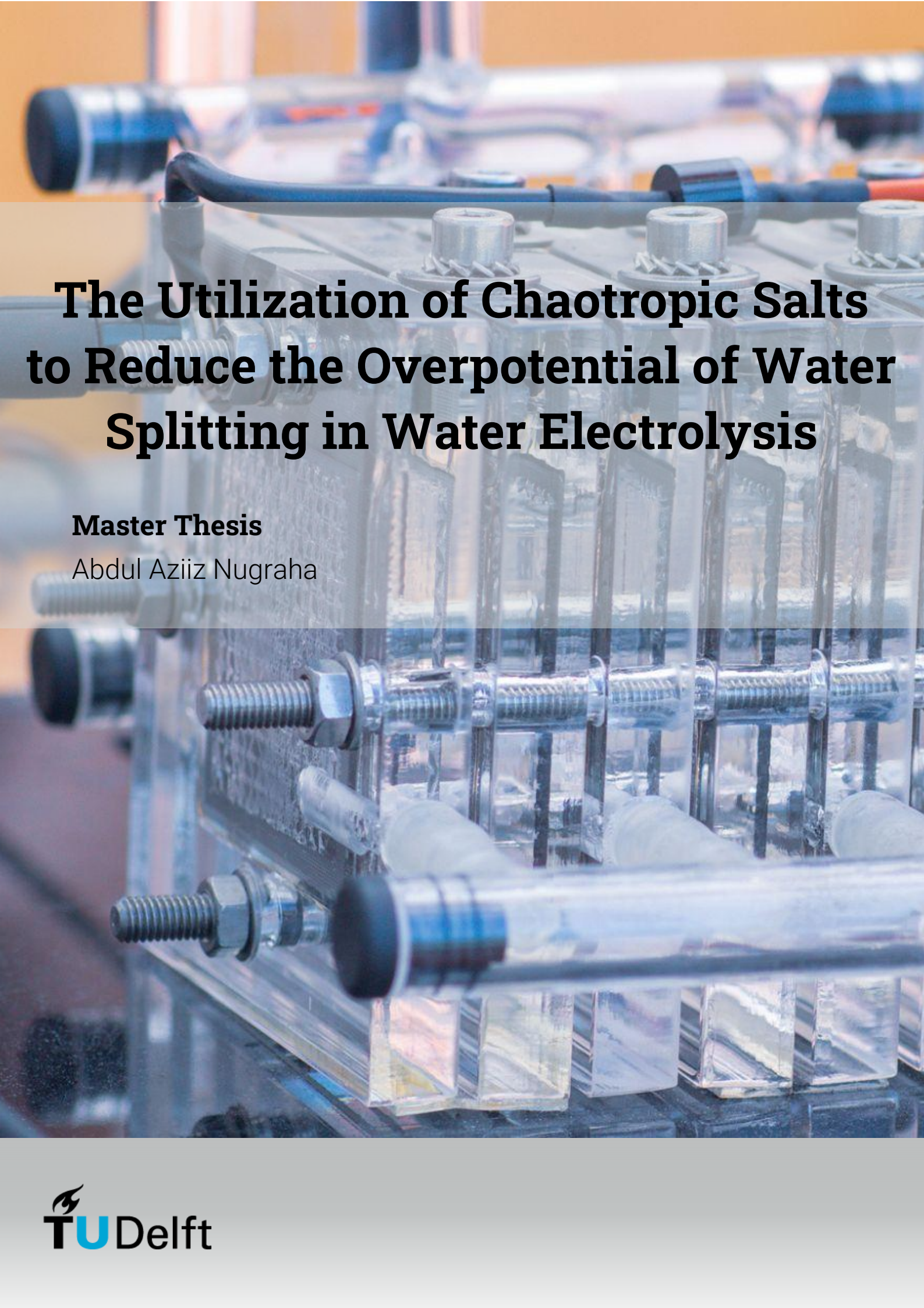




The Utilization of Chaotropic Salts to Reduce the Overpotential of Water Splitting in Water Electrolysis

Master Thesis

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The Utilization of Chaotropic Salts to Reduce the Overpotential of Water Splitting in Water Electrolysis

by

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Preface

Competing this master thesis has been a challenging yet deeply rewarding experience, building on the lessons I learned during my bachelor's graduation project. Throughout this journey, unexpected challenges—including instrument failures, demanding repetitive work, and puzzling experimental results—provided valuable opportunities to strengthen my perseverance, adaptability, and problem-solving skills. Although additional time would have allowed me to explore some of these issues more comprehensively, I am deeply grateful to have had the opportunity to undertake and complete this thesis.

I would like to express my sincere gratitude to everyone who has been part of this two-year journey. First and foremost, I thank Allah SWT, whose grace and gift of knowledge gave me the faith and strength to complete this project. Second, to LPDP, for awarding me a full scholarship and ensuring that all my financial needs were met throughout my studies. Third, I would like to thank my beloved wife and our baby for their unwavering support and the heartwarming smiles that greeted me whenever I returned home—a true source of emotional strength and motivation.

I would also like to extend my sincere appreciation to Thijs, who have supervised my work over the past nine months. His valuable feedback and insights during progress meetings have helped me overcome many difficult challenges. I am equally grateful to Mahinder and Jelle for their guidance throughout the project. I truly appreciate the time and support they devoted to providing advice and engaging in scientific discussions. And particularly to Jelle, his explanations of molecular dynamic simulations—and, more importantly, the simulation results he provided—made an essential contribution to the development of this thesis.

Lastly, my gratitude also extends to my friends in Delft, especially the members of SET TU Delft 2024, for the laughter, potluck gatherings, barbeques, beach-volleyball sessions at X.

Abdul Aziiz Nugraha
Delft, August 2026

Abstract

This study investigates the influence of sodium thiocyanate (NaSCN) as chaotropic electrolyte additive on the physicochemical properties and electrochemical performance of aqueous NaOH for alkaline water electrolysis. Density, viscosity, and electrical conductivity were measured experimentally and compared with empirical correlations and classical MD simulations. A biased-random walk model was used to estimate the contribution of Grotthuss-type transport to OH^- mobility. Electrochemical performance was evaluated on Pt-mesh electrodes using cyclic voltammetry. NaSCN addition increased electrolyte density and generally increased viscosity. Although it increased the number of charge carriers, its effect on conductivity depended strongly on composition. Positive conductivity increments were observed in some diluted mixtures, whereas concentrated mixtures exhibited reduced conductivity because of increased viscosity, ionic crowding, and correlated ion motion. Classical simulations reproduced NaSCN conductivity but underestimated NaOH conductivity because the nonreactive force field excluded structural OH^- transport. The conductivity-based analysis indicated that NaSCN decreased the corrected OH^- self-diffusion coefficient and increased its estimated hopping lifetime, suggesting suppression of Grotthuss transport. NaSCN also substantially inhibited the electrochemical reactions on Pt. The HER overpotential magnitude at -10 mA/cm^2 increased approximately from 116 mV to 300–310 mV, while the apparent OER overpotential at 10 mA/cm^2 increased approximately from 459 mV to as much as 1082 mV. These effects are attributed to a combination of reduced ionic mobility, increased transport resistance, modification of the electrical double layer, and specific adsorption of SCN^- on Pt. Sulfate detected after cyclic voltammetry further supported the occurrence of parasitic SCN^- oxidation, indicating that part of the anodic current could not be assigned exclusively to OER. Overall, NaSCN did not improve alkaline water electrolysis performance under the investigated conditions. The results demonstrate that chaotropicity alone is insufficient for selecting electrolyte additives; ionic transport, catalyst interactions, and electrochemical stability must also be considered.

1 Introduction

Green hydrogen (H_2) plays an important role as an emerging option for energy transitions and decarbonization efforts in the current energy market.^[1] It can serve as an alternative fuel in the transportation and power generation sectors, and as a clean substitute for fossil-based (grey) hydrogen in traditional applications, such as a feedstock in industrial operations.^[2] Because of its high gravimetric density, it allows for a long term energy storage to address seasonal energy mismatches.^[3] In 2024, Global Hydrogen Review reports a nearly 10% increase in low-emissions hydrogen demand compared to 2023, despite its low share (less than 1%) in global hydrogen production.^{[2], [4]} Unlike fossil-based hydrogen, green hydrogen is produced using water electrolysis powered by renewable energy.^[1] As a result, this process eliminates direct carbon output. However, the cost gap between green and grey hydrogen remains as a hinderance for a large-scale green hydrogen production.^{[4], [5]} Therefore, highly efficiency and techno-economically feasible green hydrogen production is required to narrow this gap.

Alkaline water electrolysis (AWE) stands among the most mature and established technologies to produce large-scale green hydrogen. It is also the technology with the largest manufacturing capacity at present.^{[4], [6], [7]} In alkaline electrolyzers, NaOH or KOH solutions are commonly utilized as the electrolyte and Ni-based transition metal catalysts are generally used as the electrodes.^{[6], [8]} Figure 1-1 shows a basic configuration for a typical alkaline water electrolyzer. Under standard conditions, the minimum thermodynamic reversible potential required for water splitting (based on the overall reaction) is 1.23 V. In practice, however, industrial alkaline electrolyzers typically operate at cell voltages around 1.9–2 V and at current densities of about 0.2–0.6 A/cm², with a voltage efficiency (HHV) above 70%.^[6] The higher practical operating potential arises from kinetic, ohmic, and mass transport losses within the cell. This remains one of many challenges, mainly on how to design the cell system with improved and reliable performance without losing its cost competitiveness.

Extensive research has been conducted to advance green hydrogen production technologies. Most of the studies aim to investigate the means to design a scalable green hydrogen production along with low energy consumption and economical processes, especially for AWE. For instance, several studies have been conducted by varying materials in the diaphragm/separator of the cell,^{[9], [10]} improving the activity of the catalysts,^{[11]–[13]} or optimizing the operating conditions^[14]. However, modifications to the cell components including electrode alloying or catalyst deposition pose drawbacks such as high fabrication cost, technical difficulties or scalability issues.

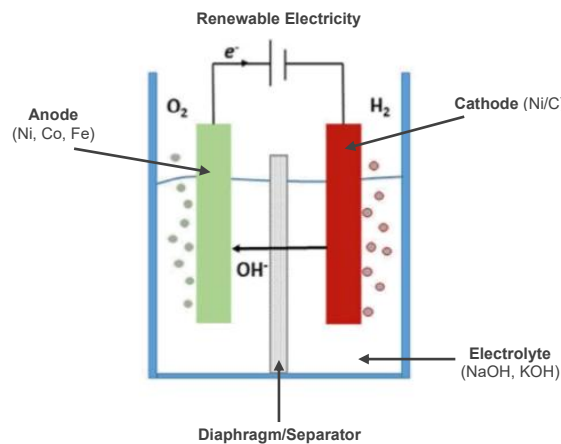


Figure 1-1 Basic Alkaline Water Electrolyzer Cell Configuration

Regardless of the improvements achieved through operating-condition optimization or cell modification, these approaches often involve complex fabrication procedures, high material costs, or scalability limitations. Therefore, increasing attention has also been directed toward electrolyte

engineering as a simpler and potentially more cost-effective strategy for enhancing AWE performance. For instance, ionic liquid (IL) additives have been extensively studied in various electrochemical applications. Their unique properties, including a wide electrochemical window, high ionic conductivity, and thermal stability, have enabled them to emerge as a promising alternative.^{[15], [16]} These studies demonstrated enhanced hydrogen production and reduced overpotential in the presence of low concentrations of ILs.^{[17]–[19]} For instance, the addition of 1 vol% of [Cho][Pro] (choline proline) amino acid ionic liquid into the electrolyte solution could increase HER reaction kinetics.^[17] This was indicated by a lower Tafel slope result compared to KOH electrolyte without ionic liquid additions. The zwitterionic (molecule that equally contains both positively and negatively charged groups) characteristic of proline helps increase ionic conductivity and proton availability, while choline may change the electric double-layer structure at the electrode–electrolyte interface, enabling proton transfer.^[17] Substantial decreases in charge transfer resistance (R_{ct}), polarization resistance (R_p) and overall impedance were observed for an electrolyte and salicylate-based ionic liquids mixtures in a low temperature AWE.^[18] Thus, previous studies have demonstrated that electrolyte engineering is able to enhance the performance of AWE.

The improvement shown with IL additives highlighted the importance of tailoring electrolyte structures and electrode–electrolyte interfacial interactions in HER kinetics. These findings suggest that controlling hydrogen-bond network of water molecules and the ions near the electrode surface may improve the electrolysis performance. Based on this concept, chaotropic agents emerge as a potential material due to their ability to disrupt the hydrogen bonding network between water molecules.^[20] One study showed that introducing a chaotropic molecular monolayer on the electrocatalyst resulted approximately a two-fold enhancement in proton transfer kinetics and three-fold reduction in HER overpotential relative to emerging electrocatalytic designs including carbon-based materials, transition metals, noble metals, and conventional surface modifications.^[21] However, this study primarily focuses on catalyst modification rather than electrolyte modification. Moreover, the electrode preparation procedure involves a relatively complex chemisorption process to incorporate the chaotropic agents into a nanoporous gold bowl (NPGb) surface which subsequently limits the economic feasibility and scalability of the electrolysis cell system. Nevertheless, the results of the study have confirmed enhanced water splitting efficiency in the presence of chaotropic agents within the electrolysis cell system. Despite the promising effects of chaotropic compounds on proton-transfer kinetics, their direct application as electrolyte additives in alkaline water electrolysis remains largely unexplored. The present study offers a less complicated method through the addition of low concentration chaotropic agents in the electrolyte solution. Furthermore, the chaotropic agents will primarily be selected from the Hofmeister series due to their availability and extensive application in the field of biology.^[22] Chaotropic agents are expected to disrupt the hydrogen-bond network of water molecules on electrode surfaces, thereby increasing the efficiency of water-splitting reaction in hydrogen generation as illustrated in Figure 1-2.

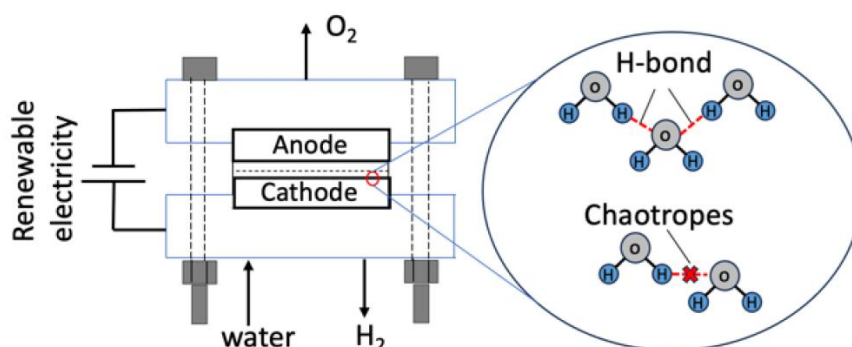


Figure 1-2 Schematic of a hydrogen electrolyzer, where water is split at the cathode to produce hydrogen, while oxygen is produced at the anode. Inset shows hydrogen-bond (H-bond) network of water, and the use of chaotropes to break these H-bonds.

This study investigates the effect of modified electrolyte mixtures on the thermophysical and transport properties as well as the electrochemical reaction overpotential following the addition of thiocyanate anion (SCN^-) to a NaOH electrolyte solution. Changes in properties such as density, viscosity, and electrical conductivity can affect mass transport and ohmic losses and, consequently, the measured electrolysis performance. Therefore, these properties are measured to distinguish direct electrochemical effects of SCN^- from indirect effects arising from the changes in bulk electrolyte. Moreover, experiments are performed to assess the electrolysis performance in various electrolyte mixtures in a specified electrolysis setup. To the best of our knowledge, limited studies have been conducted on electrolyte modification by chaotropic agents. Thus, this study mainly aims to provide new insight on the behavior and performance of a modified electrolyte solution by adding a certain concentration of chaotropic agents in the AWE and subsequently offering an alternative way to enhance the efficiency of water-splitting reaction. Moreover, the availability of chaotropic agents as materials may offer a more practical approach in enhanced water splitting reaction. Certain chaotropic agents may offer practical advantages compared to conventional IL additives due to their simpler synthesis routes and potentially reduced environmental concerns, for instance imidazolium cations are commonly used ILs as electrolyte additives, however they also hold environmental issues and toxicity, in addition to their high costs.^[17] It is also noteworthy that a small potential reduction of only 0.1 V corresponds to a total savings of approximately 111 million €/year for a big hydrogen park like the one planned in Groningen (NorthH₂ project), which aims to produce 800,000 tons of hydrogen per year in 2040.^[23]

A main research question and sub-questions are formulated to explain the scope of the study. The research questions are listed and explained as follows.

- Main research question:

How does the addition of thiocyanate (SCN^-) ions to an alkaline electrolyte influence the water electrolysis performance?

The main research question shall be answered with a few sub-questions.

1. How do the characteristics of the electrolyte change upon the addition of different concentration of thiocyanate (SCN^-) ions?

For this purpose, experimental measurements on the NaOH/NaSCN solution mixtures are conducted for several properties, including density, viscosity and electrical conductivity at several NaSCN concentrations. These measurements are used to determine whether changes in electrolysis performance originate from a direct influence of SCN^- on the electrochemical reactions or indirectly from altered bulk-electrolyte properties, such as mass transport and ohmic resistance. Molecular dynamic simulation and empirical correlation are also compared with the experimental measurements to validate the molecular models and characterize ion-transport. Specifically, the difference between experimental and classically simulated electrical conductivities is used to investigate the self-diffusion coefficient and lifetime of the charge carrier (OH^-), following the approach of Lagerweij *et al.*^[28]

2. How does the addition of thiocyanate (SCN^-) ions increase the performance of alkaline water electrolysis?

For this purpose, electrochemical experiments are conducted in a three-electrode setup to investigate electrolysis performance through actual overpotential requirements. The results are evaluated to provide comprehensive insight into the effect of thiocyanate (SCN^-) ions on the electrochemical process.

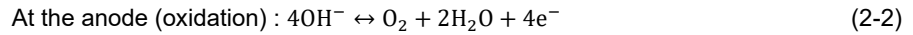
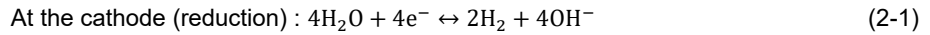
The thesis is structured as follows. Chapter 2 provides a review of the relevant literature on the chaotropic agents selection and the experiment configuration. Chapter 3 describes the methodology employed in this study. Chapter 4 presents and discusses the experimental results, including the performance of water-splitting process with and without chaotropic agent additions, together with the recommendations for future work. Finally, Chapter 5 summarizes the main conclusions of the research.

2 Literature Review

Extensive research has been conducted on the physicochemical properties of chaotropic salts, including their thermophysical properties, ion-water interactions, and influence on the hydrogen-bond network of aqueous solutions. Likewise, considerable research has focused on electrolyte and electrolyte engineering strategies to improve the performance of water electrolysis systems. However, studies investigating the application of chaotropic salts as electrolyte additives specifically for alkaline water electrolysis are limited. The limited overlap between these two research areas necessitates drawing upon studies of electrochemical behavior of chaotropic ions alongside electrolyte engineering strategies reported for water electrolysis. These studies provide the basis for assessing the suitability and stability of chaotropic additives and guide the selection of materials and methodologies employed in this work.

2.1 Fundamentals of Alkaline Water Electrolysis and Electrolyte Engineering

Alkaline water electrolysis (AWE) is one of the most mature and promising technologies for green hydrogen production. A typical AWE cell consists of a cathode, an anode, an alkaline electrolyte and an external power source. Two essential reactions occur in water electrolysis, namely hydrogen evolution reaction (HER) at cathode and oxygen evolution reaction (OER) at anode, as represented by the following equations^[24]



Water splitting reaction is a non-spontaneous process which requires a minimum amount of external electrical energy to initiate it.^{[6], [25]} The following equation shows the change in the Gibbs free energy ΔG of the electrochemical reaction

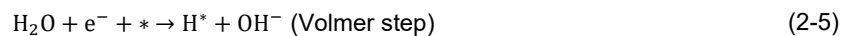
$$\Delta G^\circ = -nFE^\circ_{\text{cell}} \quad (2-3)$$

where n denotes the number of electrons transferred during the reaction, and F is the Faraday constant. Under standard conditions (25°C), the thermodynamic equilibrium cell voltage, E°_{cell} , for overall water electrolysis cell reaction is 1.23 V, corresponding to the change in the Gibbs free energy of the reaction of +237.2 kJ/mol. In practice, however, industrial electrolyzers typically operate at cell voltages between 1.8 V and 2.0 V to achieve commercially relevant current density. This additional potential arises because electrochemical reactions are kinetically limited. Consequently, an overpotential (η) above the equilibrium cell voltage is necessary to overcome activation barriers, increase the reaction rate, and compensate the effect of gas bubble formation.^[24] Moreover, additional energy is also required to drive ionic transport and overcome the ohmic resistance of the electrolyte, separator, and external electrical circuit, and as a result causes a potential drop, iR_{cell} , within the cell. Thus, the cell potential (E_{cell}) can be expressed in the following equation

$$E_{\text{cell}} = E_{\text{anode}} - E_{\text{cathode}} + \sum \eta + iR_{\text{cell}} \quad (2-4)$$

The electrochemical reaction rate in an electrolysis cell depends on the properties of the electrode surface, composition and structure of the electrolyte at the electrode-electrolyte double layer interface and the applied reaction overpotential.^[25] The influence applied overpotential on reaction kinetics can be explained by examining the mechanisms of HER and OER occurring at the electrode surface.

In alkaline media, the mechanism of hydrogen generation in cathodic HER involves reduction of water molecules which results in hydrogen gas and hydroxide ions (OH^-) productions. The reaction starts with adsorption of hydrogen on the electrode surface



where * is vacant adsorption sites on the catalyst and H* represents the adsorbed hydrogen atom on the vacant sites. The subsequent steps are hydrogen generation either by chemical desorption



or electrochemical desorption



The overpotential on the cathodic HER thus governed by the kinetics of the reaction steps on the electrode surface. Consequently, the electrocatalyst material plays a crucial role in determining the HER overpotential via its interaction with adsorbed hydrogen intermediates. If the catalyst binds hydrogen too weakly, then hydrogen adsorption becomes unfavourable. Conversely, a strong hydrogen-catalyst bonds will hinder the recombination and desorption of the hydrogen as a product. Therefore, electrocatalyst with an optimum hydrogen-binding strength generally exhibit the highest HER activity.^[6] Nickel (Ni) is among the most widely used electrode materials due to its catalytic activity, availability and affordability.^{[6], [26]} Nevertheless, platinum (Pt) remains the benchmark electrocatalyst, particularly for HER electrocatalyst, because of its high catalytic activity and low overpotential despite its high production costs.^[6]

The OER proceeds through a more complex reaction mechanisms compared to the HER. It involves four electron transfer processes. Various mechanisms have been proposed for the OER, among which the following reaction pathway is the most widely accepted^{[6], [25]}



Generally, reducing the overpotential of OER is more challenging compared to HER due to its complex pathways and irreversibility. Thus, transition metal-based electrocatalysts that possess tuneable electronic structures and accessible multiple oxidation states are considered promising materials for OER in alkaline water electrolysis.^[6]

Given the limitations in the reaction kinetics of alkaline water electrolysis, extensive research has been devoted to the development of alternative electrocatalysts with enhanced catalytic activity, stability, and economical feasibility. Strategies include the development of transition metal alloys, chalcogenides, perovskite and different kinds of modifications in electrocatalysts have been conducted to reduce the overpotential in water splitting reactions.^[6] In contrast, electrolyte engineering has received relatively limited attention. However, altering the electrolyte composition and transport properties offers considerable potential to enhance the overall efficiency of the AWE. Enhanced ionic conductivity and mass transport can reduce electrolyte resistance and thereby decrease the ohmic potential loss, iR_{cell} , at a given current density. In addition, electrolyte engineering may alter the composition and structure of the electrical double layer at the electrode-electrolyte interface, potentially influencing the HER and OER activation overpotentials, η .

Alkaline electrolytes are widely employed due to their high ionic conductivity which arises from the high concentration and mobility of ions. Aqueous potassium hydroxide (KOH) or sodium hydroxide (NaOH) are popular electrolytes used in AWE. In addition to their high conductivity, these solutions are preferred to avoid corrosion loss commonly caused by acid electrolytes.^[27] In AWE, hydroxide ions (OH⁻) serve as the primary ionic charge carriers through the electrolyte, making their mobility a key factor governing electrolyte conductivity and ionic transport resistance. Their mobility arises from the combined contributions of conventional vehicular diffusion and the faster proton-hopping (Grotthuss) mechanism.^[28] Increasing the hydroxide concentration generally enhances ionic conductivity and reduces the ohmic resistance associated with ion transport. However, an optimum electrolyte concentration is required, as excessively high hydroxide concentrations also increase electrolyte viscosity and reduce ion mobility, resulting in a trade-off between ionic conductivity and mass transport

efficiency.^{[29]–[31]} Beyond optimizing the hydroxide concentration, the electrolyte transfer properties and electrochemical performance can be improved by modification of the electrolyte composition with suitable additives.^[25]

The presence of electrolyte ions modulates the local electrical field within the electrical double layer, influences the structure and dynamics of interfacial water structure, and modulates the adsorption behavior of reaction intermediates during electrochemical reactions. Therefore, selecting the appropriate electrolyte ions provides an additional means for improving the performance of AWE besides enhancing ionic conductivity. Consequently, the activation overpotential required to drive the electrode reactions can be reduced by altering the charge-transfer kinetics of the HER and OER.^{[32]–[35]}

2.2 Chaotropic Ions and Ion–Water Interactions

During the solvation of a solute, water molecules orient and reorganize around the solute while preserving, as much as possible, the hydrogen-bonding network of bulk water. The nature of the solute ions can alter the structure and dynamics of the surrounding water molecules. Kosmotropic ions are generally small sized and it has high charge density ion that creates strong directional hydrogen-bonding or coordinative interaction, binding surrounding water molecules stronger than the hydrogen-bonds between neighboring water molecules in bulk water.^{[36], [37]} In the other hand, chaotropic agents (ions), which generally characterized by large size and small charge density, are molecules that can perturb the local hydrogen-bond network of water molecules.^{[20], [36]} Thus, given its ability, the chaotropic agents are commonly used in the field of biology by influencing physicochemical properties and interactions between proteins.^[22] Chaotropic ions will interact weakly with water and instead adsorb preferentially onto the surface of protein, increasing the solubility of the protein. The Hofmeister series defined a scale according to the ions ability to solubilize/precipitate protein, which is frequently attributed to structure-making and -breaking character of ions as illustrated in Figure 2-1.^{[38], [39]}

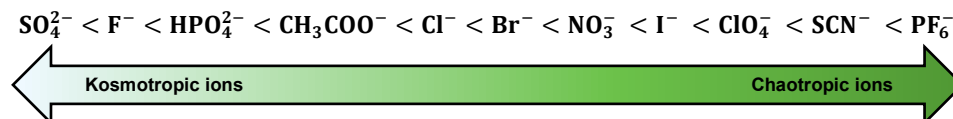


Figure 2-1 Hofmeister scale (anions) with description of the kosmotropic and chaotropic regions.

The influence of chaotropic ions on the behavior of surrounding water molecules can be evaluated through experimentally derived ion properties. Marcus developed an empirical scale of water-structural entropies of ionic hydration (ΔS_{struct} , listed in Table 2-1 for selected anions), enabling the evaluation of the kosmotropic and chaotropic ions characteristics as well as the thermochemical trends regarding their hydration processes.^[40] The decreasing state of water structure in the surrounding ion can be indicated by positive ΔS_{struct} value, this effect can also be interpreted (empirically) as an effective reduction in the total number of hydrogen bonds within the ion's solvation shell, corresponding to a negative value of ΔHB .^[40] Ions explicitly characterized as chaotropes if $\Delta HB < -1$ (or $T\Delta S_{struct} > 5$ kcal mol⁻¹).

Table 2-1 Chaotropicity scale based on the ion hydration parameters proposed by Marcus^{[37], [40]}: ion spherical radius (*r*), water-structural entropies of ions (ΔS_{struct}), and overall changes in the number of surrounding hydrogen bonds (ΔHB)

Ion	<i>r</i> [pm] ^[a]	$T\Delta S_{struct}$ [kcal mol ⁻¹] ^[b]	ΔHB ^[c]
kosmotropes			
SO ₄ ²⁻	230	-6.7	0.78
HPO ₄ ²⁻	238	-4.4	0.46
CO ₃ ²⁻	178	-4.0	0.40
F ⁻	133	-2.1	0.12
chaotropes			
Br ⁻	196	5.8	-1.01
SCN ⁻	213	5.9	-1.03
BF ₄ ⁻	230	6.6	-1.12
ClO ₄ ⁻	240	7.6	-1.27
I ⁻	220	8.3	-1.37
PF ₆ ⁻	295	8.7	-1.43 (1.93)

[a] As tabulated in [41]. [b] Calculated in accordance to Marcus [41], values for ions for which no hydration enthalpies are known were interpolated (for PF₆⁻) from a correlation of ΔS_{struct} vs. *r*. [c] From eq. 25 in [40], there abbreviated as ΔG_{HB} , it is an empirical parameter derived from hydration thermodynamics rather than a direct experimental measurement

Another descriptor commonly used to classify ions dynamic in affecting the surrounding water molecules is the B-coefficient. It originated from Jones-Dole correlation which is used to express the effect of ions on the change in dynamic viscosity of a solution.^[42] The Jones-Dole correlation is written as follows.

$$[(\eta/\eta_w^*) - 1] = Ac_2^{1/2} + Bc_2 \quad (2-12)$$

A is a constant that includes ion-ion interaction and primarily depends on coulombic interaction, this value can be calculated theoretically.^{[43], [44]} B (or B-coefficient) is an adjustable parameter related to the size of the ions. It is proportional to their partial molar entropies and can be expressed as an additive property. An ion with positive B-coefficient value (B>0, increases viscosity) is classified as water structure makers and negative B-coefficient (B<0, reduces viscosity) value represents otherwise. The value of B-coefficient for several ions is listed in Table 2-2. However, it is noteworthy to mention that the Jones-Dole equation is primarily limited to a very low concentration of solutions (<0.1 mol/kg).

Table 2-2 Jones-Dole B-coefficient properties of selected ions at 298.15 K and their effects on solution viscosity and water molecule structure; B < 0 is associated with decreased viscosity and structure-breaking characteristics, whereas B > 0 is associated with increased viscosity and structure-making behavior.^[41]

Ion	B [L/mol]	Ion	B [L/mol]
Cl ⁻	-0.005	CH ₃ CO ₂ ⁻	0.246
Br ⁻	-0.033	HCO ₃ ⁻	0.13
I ⁻	-0.073	H ₂ PO ₄ ⁻	0.34
SCN ⁻	-0.022	CO ₃ ²⁻	0.294
NO ₃ ⁻	-0.045	SO ₄ ²⁻	0.206
ClO ₄ ⁻	-0.058	HPO ₄ ²⁻	0.382

Although the thermodynamic descriptors such as ΔS_{struct} , ΔHB and B-coefficient value provide quantitative classification of chaotropic ions, they do not physically explain the molecular mechanisms governing their interaction with water molecules. Thus, understanding the molecular-scale origin of chaotropicity is essential for evaluating how chaotropic ions may influence the structure and dynamics of interfacial water, and ultimately affect the kinetics of electrochemical water splitting. Despite the debates over the subject of the traditional water structure-making and -breaking nature of various ions

among researchers, this review focuses on the experimental and computational studies describing the hydrogen-bond interactions of water molecules by the chaotropic ions from the Hofmeister series.

Based on several computational studies^{[45]–[52]} and experiments^{[53]–[56]}. Generally, these studies provide that chaotropic ions form a distinct but weakly bound first hydration shell with frequently exchanging water molecules. The hydration shell is very dynamic, and its structure is influenced more by the surrounding hydrogen-bond network rather than ion-water interactions. Subsequently, inducing a typical “structure-breaking” behavior in aqueous solution. Correlated vibrational spectroscopy (CVS) experiments by Flór *et. al.*^[53] found that chaotropic ions decrease the strength of hydrogen-bond between adjacent water molecules with elevating number of H-bonds interaction, illustrating a “structure-breaking” behavior. They also summarized a qualitative “structure-breaking” or “-making” dynamics of several anions based on their experiment results as shown in Table 2-3.

Table 2-3 Anions effect on the water hydrogen-bond network based on changes in H-bond strength and the number of correlated H-bonds. The final column summarizes the two contributions to structure-breaking or -making: the first term refers to the effect on H-bond strength, while the second refers to the effect on the number of correlated H-bonds.^[53]

anion	H-bond strength	number of correlated H-bonds	Effect on structure	
			Strength + #H-bonds ^[a]	
I [−]	↓	↑	Breaker + Maker	
SCN [−]	↓	↑	Breaker + Maker	
Br [−]	↓	↑	Breaker + Maker	
Cl [−]	↓	↑	Breaker + Maker	
ClO ₄ [−]	↓	↓	Breaker + Breaker	
NO ₃ [−]	↓	↓	Breaker + Breaker	
OH [−]	↑	↑	Maker + Maker	
F [−]	↑	-	Maker + Maker	
SO ₄ [−]	↑	↑	Maker + Maker	

Given their characteristics, chaotropic ions are expected to modify the structure and dynamics of interfacial water molecules within the electrical double layer. By weakening the local hydrogen-bond network, they may increase the configurational flexibility of water molecules, which further may facilitate water reorganization. Consequently, chaotropic ions may alter the interfacial reaction environment and influence the charge-transfer kinetics of AWE.

2.3 Analyzing Chaotropic Ion Electrochemical Stability

In electrolyte engineering, suitable additives should not only enhance the physicochemical properties of the electrolyte but also possess sufficient electrochemical and chemical stability under the intended operating conditions. They should not undergo parasitic side reactions within the potential ranges required for HER and OER or generate products that interfere with either electrode reaction. Additive stability should be evaluated under the practical working potential range of the complete alkaline water electrolysis system.^[57] Based on the required oxidation potential, some anions could maintain their electrochemical stability within the range of OER onset potential; such as F[−] and SCN[−]. An illustration for several anions standard electrode potential is shown in Figure 2-2.

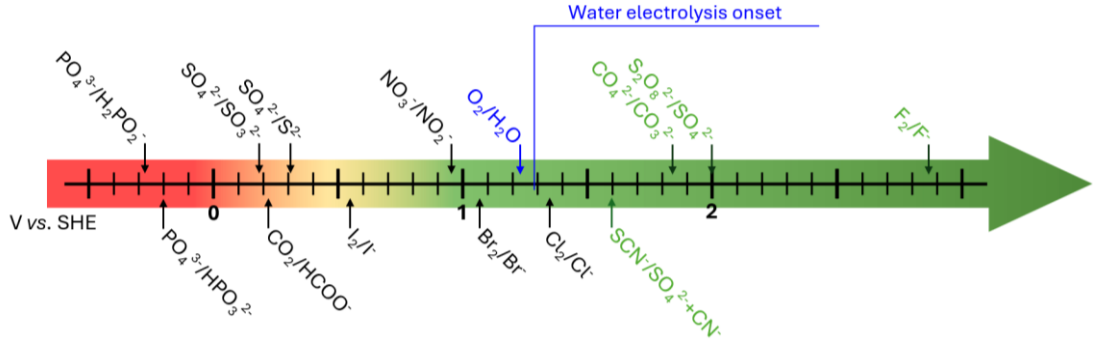


Figure 2-2 Standard electrode voltage for several anions

Regarding chemical stability in aqueous electrolyte, anions such as PF_6^- and BF_4^- are widely employed in non-aqueous solvent (such as ionic liquid or organic media) electrolytes due to their high electrochemical stability, allowing electrolyte systems to operate over a wide electrochemical range (~ 0 – 5 V vs. RHE). However, these anions are susceptible to hydrolysis in aqueous media, specifically under acidic conditions or at elevated temperatures, resulting in formation of hydrofluoric acid (HF).^[58] Specific adsorption of electrolyte anions (e.g., SO_4^{2-} on Pt) can occupy surface active sites, competing with reactants and reaction intermediates which consequently altering the reaction pathways.^[57]

2.4 Electrolyte Mixture Characterization

Characterization of the physicochemical and transport properties of electrolyte is essential prior to electrochemical analysis. It provides fundamental insight into the bulk properties of the electrolyte that govern ionic transport and influence electrochemical performance of alkaline water electrolysis. In addition to electrode reaction kinetics, electrochemical performance is also influenced by the ability of the electrolyte to transport ions, minimize ohmic losses and maintain efficient mass transport.^[25] In alkaline water electrolysis, the hydroxide ions play an important role as the primary charge carriers in the electrolyte. They primarily determine the conductivity of the solution due to their high mobility because of proton hopping (also referred to as Grotthuss transfer),^[28] they also participate as reactants during oxygen evolution reaction (OER).^[6] The addition of chaotropic agent introduces additional ions, altering the interactions among ions and water molecules in the electrolyte solution. Consequently, the properties of the electrolyte, including conductivity, viscosity, ion mobility, and the hydrogen-bond network may change, thereby influencing both bulk ionic transport and electrode-electrolyte interfacial structure.^{[40], [60], [61]} While electrolyte properties can be determined experimentally, empirical correlations and molecular simulations provide complementary approaches for evaluating and validating the experimental measurements and improving the understanding of electrolyte behavior.

Laliberté and Cooper developed an empirical model to predict density of aqueous solutions containing one or more electrolytes.^[62] Experimental density data for water-electrolyte solutions are first used to determine the apparent specific volume, $\bar{v}_{app,i}$, of electrolyte i . In contrast to an ideal-mixture model, the apparent specific volume represents both the influence of temperature and concentration on the electrolyte. The equation for calculating the apparent specific volume of one electrolyte in solution with water is expressed as follows

$$\bar{v}_{app,i} = \frac{1 - \frac{\rho_m(1 - w_i)}{\rho_{H_2O}}}{\rho_m w_i} \quad (2-13)$$

Where ρ_m is the density of the mixture in kg/m^3 , w_i is the mass fraction of component i ($w_i = 1 - w_{H_2O}$), ρ_{H_2O} is the density of water in kg/m^3 , and apparent specific volume ($\bar{v}_{app,i}$) is expressed as volume per unit mass of electrolyte i (m^3/kg solute). The correlation from Kell^[63] is used to calculate the density of water, ρ_{H_2O} , as a function of temperature.

Laliberté and Cooper found that the following correlation adequately represented the apparent specific volumes of the electrolytes considered

$$\bar{v}_{app,i} = \frac{w_i + c_2 + c_3 t}{(c_0 w_i + c_1) e^{(10^{-6}(t+c_4)^2)}} \quad (2-14)$$

where t is temperature in °C and c_0 to c_4 are electrolyte-specific empirical constants. The constants c_0 and c_1 are expressed in kg/m³, c_2 is dimensionless, c_3 has units of °C⁻¹, and c_4 is expressed in °C. For a set of trial constants, Equation (2-14) is evaluated at each experimental temperature and composition. The corresponding solution density is then calculated from

$$\rho_{m,calc} = \frac{1}{w_{H_2O} \bar{v}_{H_2O} + \sum_i w_i \bar{v}_{app,i}} \quad (2-15)$$

where $\bar{v}_{H_2O} = 1/\rho_{H_2O}$. For every experimental point i , the residual density is defined as

$$r_i = \rho_{m,exp,i} - \rho_{m,calc,i} \quad (2-16)$$

a nonlinear least-squares method is used to adjust the empirical constants, c_0 to c_4 , by minimizing the sum of the square of the residuals. Once fitted, density of the solutions can be predicted at other temperatures and mass fractions. Interpolated predictions are generally most reliable, while extrapolated results should be interpreted cautiously as their uncertainty increases with distance from the fitted temperature and concentration ranges. They also found that Equation (2-14) and (2-15) provided adequate predictions for multi-electrolyte solutions at low total electrolyte concentrations. At higher concentration, however, the prediction error increased. The accuracy is significantly improved by modifying Equation (2-14) to use the total electrolyte mass fraction ($1 - w_{H_2O}$), instead of the individual electrolyte mass fraction (w_i), when calculating the electrolyte apparent specific volume. The modified correlation is

$$\bar{v}_{app,i} = \frac{(1 - w_{H_2O}) + c_2 + c_3 t}{(c_0(1 - w_{H_2O}) + c_1) e^{(10^{-6}(t+c_4)^2)}} \quad (2-17)$$

this term represents the total mass fraction of all electrolytes in the solution. For a solution containing only one electrolyte, the modified correlation reduces to Equation (2-14).

Laliberté also developed an empirical model for predicting dynamic viscosity of aqueous solutions containing one or more solutes.^[61] Equation (2-18) is used to calculate the solute viscosity, η_i , in a single electrolyte in water

$$\eta_i = \left(\frac{\eta_m}{\eta_w^{w_w}} \right)^{1/w_i} \quad (2-18)$$

where η_m and η_w are the solution viscosity (which is obtained from experimental data) and the water viscosity, both in mPa·s, while w_i and w_w are the solute and water mass fraction, respectively. The following equations are used to calculate the viscosity of pure water

$$\eta_w = \frac{t + 246}{(0.05594t + 5.2842)t + 137.37} \quad (2-19)$$

where t is temperature and has a unit of °C. The following correlation was developed and found to sufficiently represent the viscosities of solutes

$$\eta_i = \exp \left(\frac{v_1(1 - w_w)^{v_2} + v_3}{(v_4 t + 1)(v_5(1 - w_w)^{v_6} + 1)} \right) \quad (2-20)$$

This equation is also suitable to predict the viscosity of multi-electrolyte solutions. Subsequently, he used a similar methodology to predict the density of electrolytes^[62]

1. Enter initial values for the empirical constants v_1 to v_6 .
2. Calculate the residual viscosity, δ_i , from the subtraction of experimental viscosity and calculated value ($\delta_i = \eta_{exp,i} - \eta_{calc,i}$).
3. Minimize the sum of the square of the residuals by varying v_1 to v_6 .
4. Check the consistency of the calculations. Inconsistent data will be removed and repeat steps 2 and 3 until the data is consistent.

Comparable to the empirical density model, the dynamic viscosity model produced predictions that agreed closely with experimental data when interpolation is performed within the fitted composition range. In the other hand, extrapolation substantially beyond the maximum fitted solute mass fraction led to rapidly increasing errors. Although, for instance in the CaCl_2 test case, extrapolation of approximately 10-20% beyond the fitted concentration range may be acceptable, predictions outside this range should be interpreted cautiously.

In addition to empirical model for predicting the thermophysical properties of the aqueous electrolytes, molecular dynamic simulation is also introduced to investigate their molecular-level structure and transport behavior. The study by Lagerweij *et al.* demonstrated that the transport of hydroxide (and hydronium) ions, for which self-diffusion coefficient and electrical conductivity are fundamentally governed by vehicular transport and the structural transport mechanism.^[28] Because classical molecular dynamics simulations capture only vehicular transport, the authors supplemented the classical trajectories with a biased random walk model. Assuming that there is no correlation between the vehicular and structural trajectories, the authors expressed the combined electrical conductivity, σ^{C+BRW} , as

$$\sigma^{C+BRW} = \sigma^C + \frac{e^2 z^2 N_{\text{Grott-ion}}}{k_B T V} \frac{n - nq}{n + nq - 2} \frac{\Delta_{\text{hop}}^2}{6\tau} \quad (2-21)$$

The first term, σ^C , represents electrical conductivity due to vehicular transport. it is calculated using the Einstein-Helfand equation, in which the conductivity is expressed through Onsager transport coefficients derived from the MD trajectories. The second term in Equation (2-21) represents the structural contribution associated with Grotthuss transfer. Here, e represents the electron charge, z is the absolute ionic charge number, k_B is the Boltzmann constant, T is the temperature of the system, V is the simulation-box volume, and $N_{\text{Grott-ion}}$ are the number of ions that undergo Grotthuss transfer; which in alkaline medium refers to OH^- ions. The remaining prefactor constitutes the biased random-walk contribution to self-diffusion coefficient. Under the assumptions of the model, the combined electrical conductivity, σ^{C+BRW} , should also represent electrical conductivity from experimental measurements, which naturally includes vehicular and structural transport.

Lagerweij *et al.* also proposed an approach for calculating the self-diffusion coefficient by combining the vehicular and structural trajectories of the ions.^[28] Assuming that the vehicular and structural trajectories are uncorrelated, they derived the combined self-diffusion coefficient, $D_{\text{self}_i}^{C+BRW}$, as

$$D_{\text{self}_i}^{C+BRW} = D_{\text{self}_i}^C + \frac{n - nq}{n + nq - 2} \frac{\Delta_{\text{hop}}^2}{6\tau} \quad (2-22)$$

Here, $D_{\text{self}_i}^C$ is the vehicular contribution obtained from the mean squared displacements (MSD) of species i using the Einstein equation. Because this contribution is calculated from classical MD trajectories generated in a cubic simulation box with periodic boundary conditions, the Yeh–Hummer correction is applied to account for finite-size effects.

The second term in Equation (2-22) represent the structural trajectory contribution described by the biased random walk. In contrast to the classical vehicular trajectory, the biased random walk expression is derived for an infinite system size, no finite-size correction was applied to this term. The parameter n represents the number of possible directions available for a Grotthuss proton-transfer event, while q denotes the probability of that ion hops back to its previous site. For an unbiased random walk, all transfer directions are equally probable ($q = 1/n$), while higher values of q represent an increasing bias toward back-hopping, reducing the effective structural diffusion coefficients. Finally, τ and Δ_{hop} are the expression from the self-diffusion coefficient of a simple random walk, where they denote the mean lifetime of the Grotthuss-transferring ion and the average displacement associated with one transfer event, respectively.

By setting the σ^{C+BRW} equals to the experimentally measured conductivity, σ^{exp} , the mean lifetime, τ , of a Grotthuss transferring ion can be obtained from the difference between the experimental conductivity (σ^{exp}) and the conductivity calculated using classical MD (σ^C)

$$\tau = \frac{n - nq}{n + nq - 2} \frac{\Delta_{hop}^2}{6} \frac{e^2 z^2 N_{Grott-ion}}{k_B TV (\sigma^{exp} - \sigma^C)} \quad (2-23)$$

Subsequently, substituting τ from Equation (2-23) into Equation (2-22) gives the total self-diffusion coefficient of the Grotthuss-transferring ion species; such as OH^- , ($D_{self,Grott-ion}^{C+BRW}$). This expression demonstrates that the total self-diffusion coefficient is independent of both the Grotthuss transfer distance (Δ_{hop}) and the back-hopping bias (q)

$$D_{self,Grott-ion}^{C+BRW} = D_{self,Grott-ion}^C + \frac{k_B TV (\sigma^{exp} - \sigma^C)}{e^2 z^2 N_{Grott-ion}} \quad (2-24)$$

thus, the difference $\sigma^{exp} - \sigma^C$ is attributed to structural transport through the Grotthuss mechanism.

Before electrochemical measurements are performed, empirical modeling, molecular simulations, and physicochemical characterization can be used to assess the bulk properties and transport behavior of the electrolyte mixtures. Relating these properties to subsequent HER and OER measurements may help identify the factors governing electrochemical performance and guide the selection of suitable additives for electrolyte engineering.

3 Methodology

The solutions were prepared using deionized water produced by a Milli-Q® IQ 7000 (Sigma-Aldrich, resistivity of approximately 18.2 MΩ.cm and TOC < 2.5 ppb). All chemicals were used as received without further purification: reagent-grade sodium hydroxide (Sigma-Aldrich, ≥98% anhydrous pellets) and semiconductor-grade sodium thiocyanate (Sigma-Aldrich, ≥99.99% trace metals basis).

The chemicals were used to prepare three groups of electrolyte solutions: (1) NaOH aqueous solutions (0.2–8 mol/kg H₂O), (2) NaSCN aqueous solutions (0.5–6 mol/kg H₂O), and (3) mixed NaOH-NaSCN electrolytes consisting of NaOH concentration ranging from 0.5 to 6 mol/kg H₂O and NaSCN additions ranging from 0.1 to 1 mol/kg H₂O. Mixed NaOH-NaSCN electrolytes were prepared by first dissolving the required amount and concentration of NaOH in deionized water, followed by the addition of NaSCN to achieve the desired molality. All concentrations were prepared on a molality basis by gravimetric weighing of the required masses of water and solutes. The electrolyte solutions were prepared at ambient laboratory temperature (approximately 22–25 °C) and stirred using magnetic stirrer to complete dissolution prior to measurements.

3.1 Material Characterizations

3.1.1 Density

Density measurements were performed using an oscillating U-tube density meter DMA 5002 (Anton Paar) with precision of ±0.00001 g/cm³. The instrument was equipped with a borosilicate glass tube, and sample viscosity was automatically corrected. The presented experimental density values are obtained as an average of at least three measurements at 20°C. The standard uncertainty of the density measurement was below 6×10^{-4} g/cm³.

A density correlation proposed by Laliberté and Cooper^[62] (see section 2.4) was used to calculate the densities of aqueous solutions containing NaSCN as single and multi-solute. The predicted values were subsequently compared with the experimental measurements to assess the agreement between the calculated and measured densities. Furthermore, densities predicted by classical molecular dynamics (MD) simulations were included to enable a comprehensive comparison between the empirical correlation, simulation results, and experimental measurements.

3.1.2 Viscosity

Viscosity measurements were conducted using a Modular Compact Rheometer (MCR 302, Anton Paar) with integrated temperature control system. A parallel-plate type measuring system (PP50, D: 50mm) was selected for its suitability for low-viscosity electrolyte solutions. Shear-rate-controlled rotational measurements were conducted over a shear-rate range of 150–0.5 s⁻¹, with a total of 21 data points collected for each measurement. Viscosity was calculated as the average data points where the measured viscosity reached a stable plateau (between 63.7 s⁻¹ and 4.9 s⁻¹ region). Reported experimental values represent the average of three independent measurements conducted at 20°C. The measurement uncertainty was found at below 9×10^{-5} Pa·s.

The experimental viscosity data were evaluated against the correlation proposed by Laliberté^[61] (see section 2.4). The correlation was also used to calculate the viscosity of single and multi-electrolyte NaSCN aqueous solutions. In addition, viscosity values predicted from classical MD simulations were incorporated to provide additional basis for comparison with the experimental data.

3.1.3 Electric Conductivity

Electrical conductivity measurements at 20°C was performed using a SevenCompact™ S230 conductivity meter (Mettler Toledo) equipped with Cond probe InLab 731 probe. The instrument was

calibrated at 20°C before measurements. The provided experimental values are the average of three measurements. The standard uncertainty of the measured electrical conductivity was below 0.45 S/m.

To evaluate the agreement with the experimental results, electrical conductivity was also predicted using classical MD simulations. The predicted conductivity accounts for both the vehicular transport from the classical MD trajectories and the structural (Grotthuss) transport modeled using a biased random walk. Details of the computational methodology for calculating electrical conductivity are described in section 2.4.^[28]

3.1.4 Self-diffusion Coefficient

The self-diffusion coefficients were evaluated from classical MD simulations using the Einstein relation based on the mean squared displacement (MSD) of each species. To account for finite-size effects arising from the use of periodic boundary conditions, the Yeh-Hummer correction was applied to the calculated diffusion coefficients. As classical MD simulations describe only the vehicular transport of ions, a biased random walk model was incorporated to account for the structural transport associated with Grotthuss proton-transfer. Assuming that the vehicular and structural transport mechanisms are independent, the total self-diffusion coefficient was obtained as the sum of the classical MD and the structural transport contributions.^[28] Theoretical background of the self-diffusion coefficients is presented in section 2.4.

3.1.5 Lifetime

The mean lifetime of a Grotthuss transferring ion is inferred from the difference between the experimentally measured electrical conductivity and the vehicular contribution calculated using classical MD. The theoretical basis and calculation procedure are presented in section 2.4.

3.1.6 Simulation Details

Classical MD simulations were performed using LAMMPS (version August 2024) with the OCTP plugin to calculate the relevant transport properties, including viscosity (η), self-diffusion coefficients ($D_{self,i}^C$), and Onsager transport coefficients ($A_{i,j}^C$). Initial configurations were generated using PACKMOL (version 21.2.3). The simulations of NaOH(aq) and NaSCN(aq) contained in 1100 water molecules together with the appropriate number of ion pairs corresponding to the investigated molalities and were modeled using the classical Madrid-Transport and the DFF/OH⁻. Because no suitable SCN⁻ force-field parameters were available and compatible with the selected molecular models and transport-property framework, SCN⁻ parameters were developed specifically for this project. The SHAKE algorithm was employed to constrain the geometry of H₂O and OH⁻ throughout the simulations. Long range electrostatic interactions were treated using the particle-particle particle-mesh (PPPM) method with a relative error of 10⁻⁴. To determine the average volume of the simulation box, 10 ns simulations were first performed in the *NPT* ensemble. Subsequently, production simulations of 200 ns were conducted in the *NVT* ensemble to calculate the self-diffusion coefficients, viscosities, and Onsager coefficients. The simulations were carried out at 1 atm and 20°C using a cubic simulation box with periodic boundary conditions applied in all directions. A 2 fs time step was employed with the velocity-Verlet integration scheme, while the Nosé-Hoover thermostat and barostat used coupling constant of 0.1 and 1 ps, respectively.^[28]

The molecular dynamic simulations and force-field parameterization were not conducted personally by the author. Nevertheless, the simulation results form an integral part of this thesis as they provide the molecular level and transport-property framework required to interpret the experimental density, viscosity, conductivity, and electrochemical results. The simulation methodology is therefore reported here in sufficient detail to support the reproducibility and critical evaluation of the combined experimental-computational analysis.

3.2 Electrochemical Measurements

A three-electrode cell configuration was used for electrochemical measurements. Platinum mesh electrodes (1×1 cm), mounted in PTFE holders, were used as the working and counter electrodes. A HydroFlex® reversible hydrogen electrode (RHE) was used as the reference electrode. The platinum mesh electrodes were ultrasonically cleaned in acetone, followed by isopropanol and finished with ultrapure water before use. The distance between electrodes was maintained constant throughout all measurements to ensure consistent cell configuration and minimize variations in uncompensated solution resistance. A schematic illustration of the three-electrode cell is shown in Figure 3-1.

Alkaline electrolytes consisting of 1 mol/kg H₂O NaOH with NaSCN additions ranging from 0.1 to 1 mol/kg H₂O were investigated. The measurements were conducted in a 100 mL cell at 25°C. The electrochemical cell was immersed in a thermostatically controlled water bath to maintain the desired operating temperature.

Electrochemical measurement was carried out using a Bio-Logic SP-200 potentiostat. Cyclic voltammetry (CV) was performed over a potential range from -0.8 V to 3.0 V versus RHE to evaluate the electrochemical performance of the electrolyte mixture. CV measurements were performed at scan rate of 20 mV/s. An Open-Circuit Potential (OCP) measurement was conducted for 1 hour prior to CV. Twenty consecutive CV cycles were performed to ensure electrode stabilization and the voltammogram from the final cycle was used for analysis. Following the electrochemical measurements, the electrolyte was collected and analyzed by ionic chromatography (IC, Thermo Fisher Scientific) using a Dionex IonPac AS22 column. The analysis was performed to detect anionic species, particularly sulfate, that could indicate the occurrence of undesired side reactions during electrochemical operation.

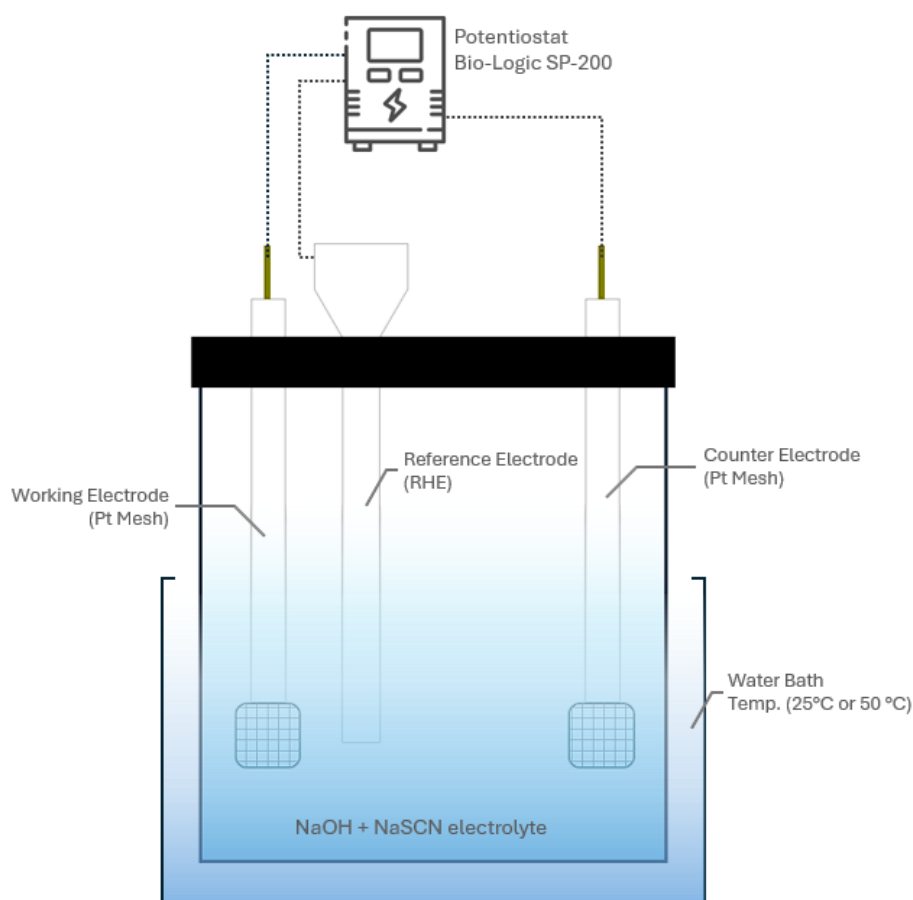


Figure 3-1 Three-electrodes cell configuration for electrolyte engineering using electrolyte mixture of NaOH and NaSCN

4 Results and Discussions

4.1 Material Characterization

Experimental measurements, prediction based on empirical correlations, and classical molecular dynamics (MD) simulations have been performed to evaluate the electrolyte properties before electrochemical analysis. The details of all the material characterizations are presented in Appendix A. For both aqueous NaOH and NaSCN solutions, the density increased with increasing molality as shown in Figure 4-1a and Figure 4-1b. The densities predicted using the Laliberté^[62] correlation and MD simulations agree well with the experimental measurements over the investigated concentration range. This behavior is expected because increasing the salt concentration introduces additional ionic mass while the corresponding increase in solution volume is comparatively smaller.

Figure 4-1c shows the excess density associated with adding NaSCN to aqueous NaOH solutions. The addition of NaSCN produced a positive density increment, which increased the NaSCN molality. However, this increment generally decreased as the NaOH concentration increased. The MD simulations closely reproduce the experimental measurements across the investigated compositions, supporting the suitability of the parametrized force field for predicting the densities of these electrolyte solutions.

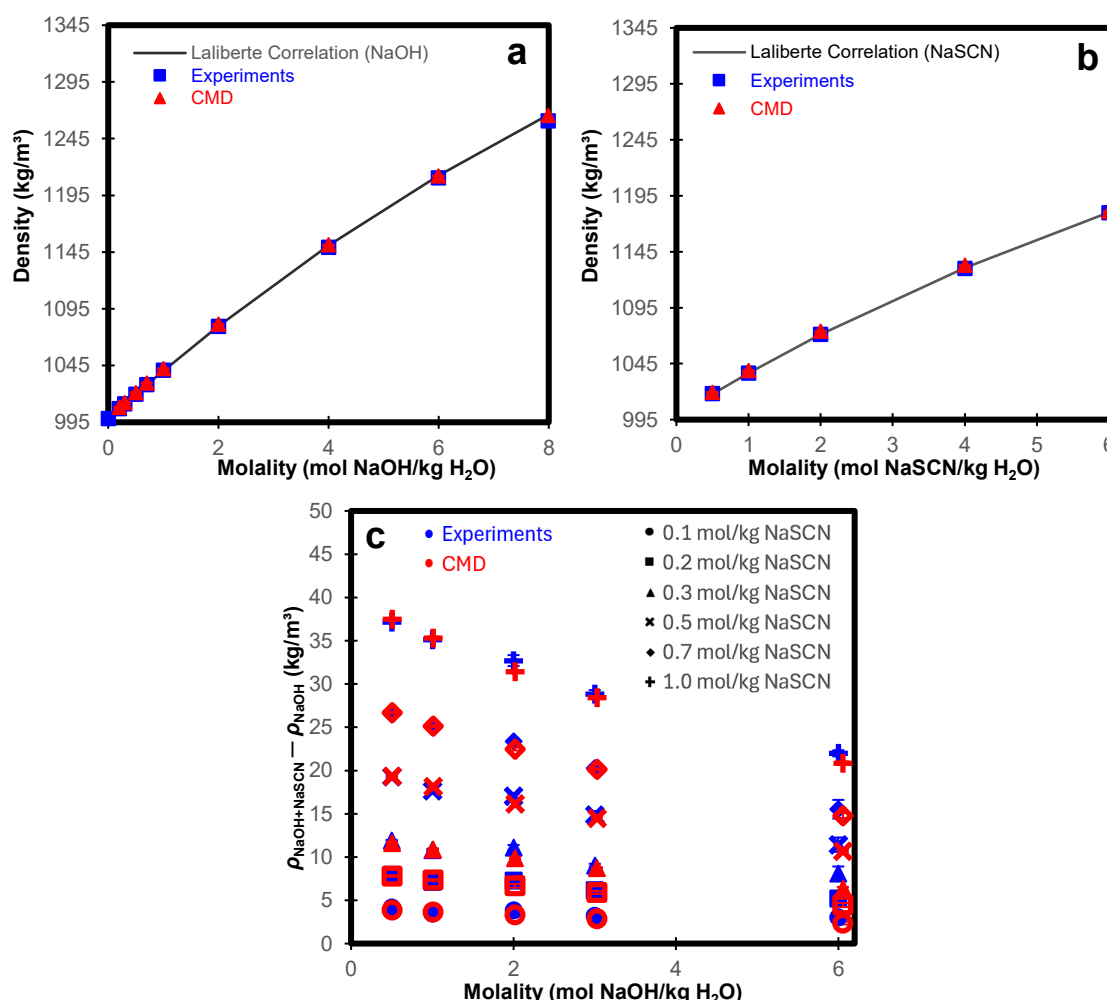


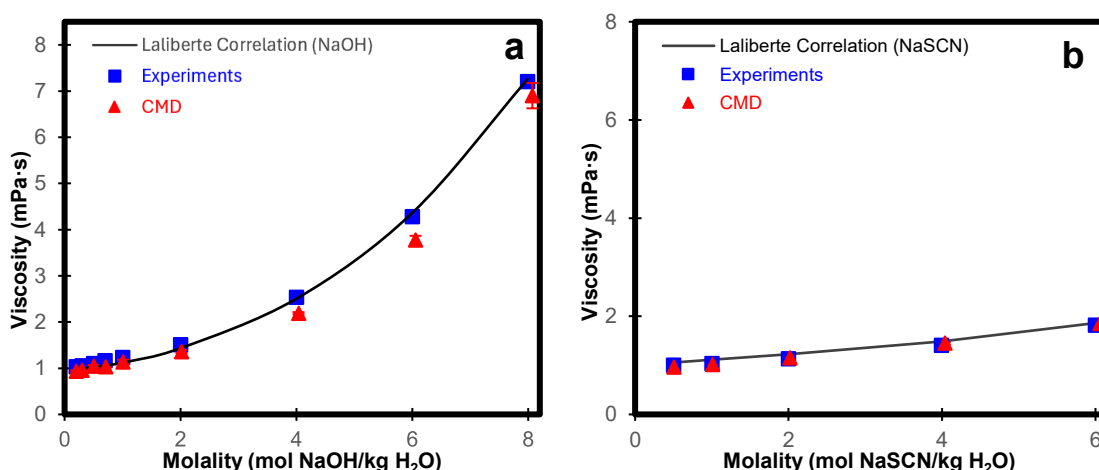
Figure 4-1 Densities of aqueous NaOH (a), NaSCN (b) solutions as functions of concentration, and the excess density due to NaSCN addition into aqueous NaOH solutions (c). The results show that the addition

of NaSCN increases the overall density of the mixtures, with the effect becoming less pronounced as the NaOH concentration increases. Overall, the CMD simulated densities (red) agree with the experimental measurements (blue) within the uncertainties, indicating that the parametrized force field accurately reproduces the density of the investigated electrolyte mixtures.

Similar evaluations are carried out for the viscosity of the electrolytes. As shown in Figure 4-2a, the viscosity of aqueous NaOH increases nonlinearly with increasing concentration. The increase becomes particularly pronounced above approximately 4 mol/kg water, indicating progressively stronger concentration-dependent interactions. The viscosities predicted using Laliberté^[61] correlation closely follow the experimental data across the investigated NaOH molality range. Meanwhile, the MD simulations reproduce similar trend but slightly underestimate the experimental viscosity at some intermediate and high NaOH molalities.

In contrast, the viscosity of aqueous NaSCN increases more gradually with increasing molality (Figure 4-2b), rising from 1.0 mPa·s at low concentration to about 1.82 mPa·s at 6 mol/kg. The nearly linear increase suggests that NaSCN has a weaker concentration-dependent effect on the liquid dynamics than NaOH over the investigated range. The simulations and the Laliberté^[61] correlation both agree closely with the experimental measurements, indicating that the force field suitably represents the viscosity of the aqueous NaSCN system.

Figure 4-2c presents the viscosity increment associated with adding NaSCN to aqueous NaOH solutions. In general, the addition of NaSCN increases the viscosity of the mixtures, and this contribution becomes larger as both the NaSCN and NaOH molalities increase. The effect is relatively small in dilute NaOH solutions but becomes more pronounced at 6 mol/kg NaOH mixtures. The small negative viscosity increments observed in dilute NaOH mixtures are consistent with the structure-breaking character of SCN^- , as indicated by its negative Jones-Dole B -coefficient.^[41] At low concentrations, SCN^- may disrupt the local hydrogen-bond network of water and increase molecular mobility, producing a small reduction in viscosity. As the NaOH and NaSCN concentrations increase, this effect becomes increasingly insignificant as ionic crowding and ion-ion and ion-water correlations progressively restrict molecular motion, leading to positive viscosity increments.^[42] The MD simulations agree reasonably well with the experimental measurements, particularly at low and moderate electrolyte mixture concentrations, supporting the suitability of the parameterized force field for predicting viscosity of these solutions. However, it is also noteworthy to mention that the deviations observed for highly concentrated ternary mixtures indicate that the ability of the force field to represent viscosity is somewhat less accurate than its performance for density.



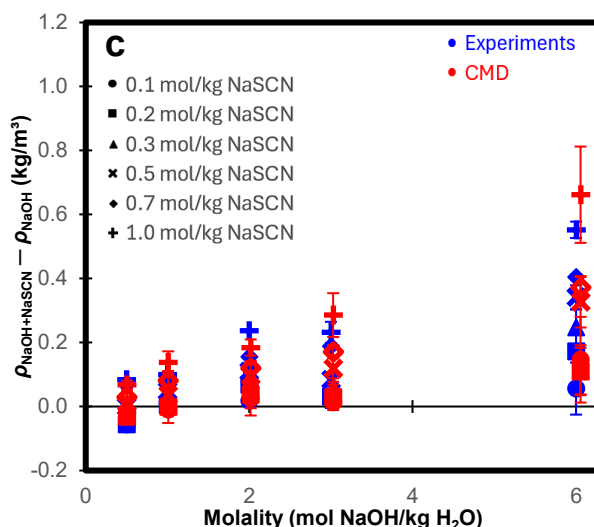


Figure 4-2 Viscosities of aqueous NaOH (a), NaSCN (b) solutions as functions of concentration, and the excess viscosity due to NaSCN addition into aqueous NaOH solutions (c). The results show that the addition of NaSCN increases the overall viscosity of the mixtures, with the effect becoming more pronounced as the NaOH concentration increases. Overall, the CMD simulated viscosities (red) agree with the experimental measurements (blue) within the uncertainties, indicating that the parametrized force field accurately reproduces the viscosity of the investigated electrolyte mixtures.

The electrical conductivities of aqueous NaOH and NaSCN solutions obtained experimentally and from classical MD simulations are shown in Figure 4-3a and Figure 4-3b. The experimental conductivity of aqueous NaOH initially increases strongly with molality, reaches a maximum at 4 mol/kg water, and subsequently decreases at higher concentrations. From 0.2–4 mol/kg water of aqueous NaOH solutions, the increasing number of charge carriers dominates the conductivity response. Beyond that, however, along with the increasing in viscosity, reduction in solvent availability and stronger ion–ion correlations increasingly restrict ionic mobility.

The CMD simulations reproduced the concentration-dependence trend of aqueous NaOH but substantially underestimate its conductivity over most of the investigated range. This discrepancy is expected due to the employed nonreactive force field which describes only the vehicular contribution to hydroxide transport. In aqueous alkaline solutions, hydroxide transport also occurs through structural trajectories, in which proton transfer between OH^- and neighboring water molecules effectively displaces the hydroxide charge through the hydrogen-bond network. This Grotthuss-type contribution is not represented by a conventional fixed-topology force field because chemical bonds cannot break or form. Consequently, the calculated conductivity primarily represents the vehicular transport and should be interpreted as a partial, rather than total, conductivity. Furthermore, as demonstrated in the work by Lagerweij *et al.*, this discrepancy between the experimentally measured conductivity and predicted conductivity from CMD directly represents the structural conductivity contributed by the Grotthuss mechanism.^[28]

The conductivity of aqueous NaSCN increases continuously over the investigated range, reaching 201.66 mS/cm at 6 mol/kg (Figure 4-3b). The close agreement between CMD and experimental results indicates that charge transport in aqueous NaSCN is predominantly vehicular and thereby adequately represented by the nonreactive force field. The gradual reduction in the slope at high molality suggests that increasing viscosity and ionic correlations begin to offset the conductivity gained from the additional charge carriers.

Figure 4-3c shows the conductivity increment associated with adding NaSCN to aqueous NaOH. Based on the experimental results, in dilute NaOH solutions, NaSCN generally gives a positive conductivity increment because it introduces additional mobile Na^+ and SCN^- ions while the solution viscosity remains relatively low. As the NaOH molality increases, the contribution becomes increasingly

smaller and eventually negative. In concentrated mixtures, the increase in charge-carrier concentration is outweighed by reduced ionic mobility resulting in higher viscosity, ionic crowding, overlapping hydration environments, and enhanced interactions among Na^+ , OH^- , and SCN^- .

The comparison of experimental and CMD simulation results at 3 mol/kg water of NaOH mixtures is particularly informative. The experimental conductivity increments are generally negative, whereas the CMD simulations predict positive increments along with increasing of added NaSCN concentrations. A plausible interpretation is that the CMD simulations capture the additional vehicular conductivity introduced by NaSCN but do not describe any disruption or modification of the proton-transfer contribution to OH^- transport. Experimentally, SCN^- may alter the water hydrogen-bond network and reduce the efficiency of hydrogen proton-transfer pathways. If this loss of structural conductivity exceeds the additional vehicular contribution from Na^+ and SCN^- , the net measured conductivity increment becomes negative.

At 6 mol/kg water of NaOH electrolyte mixtures, both experimental and CMD results show increasingly negative increments with increasing NaSCN content. Under these concentrated conditions, water availability is limited and the mixture exhibits higher viscosity and stronger ionic correlations. The negative increment predicted by CMD demonstrates that suppressed vehicular mobility alone contributes substantially to the conductivity reduction. The larger experimental decrease may additionally contain a reduction in hydroxide structural transport.

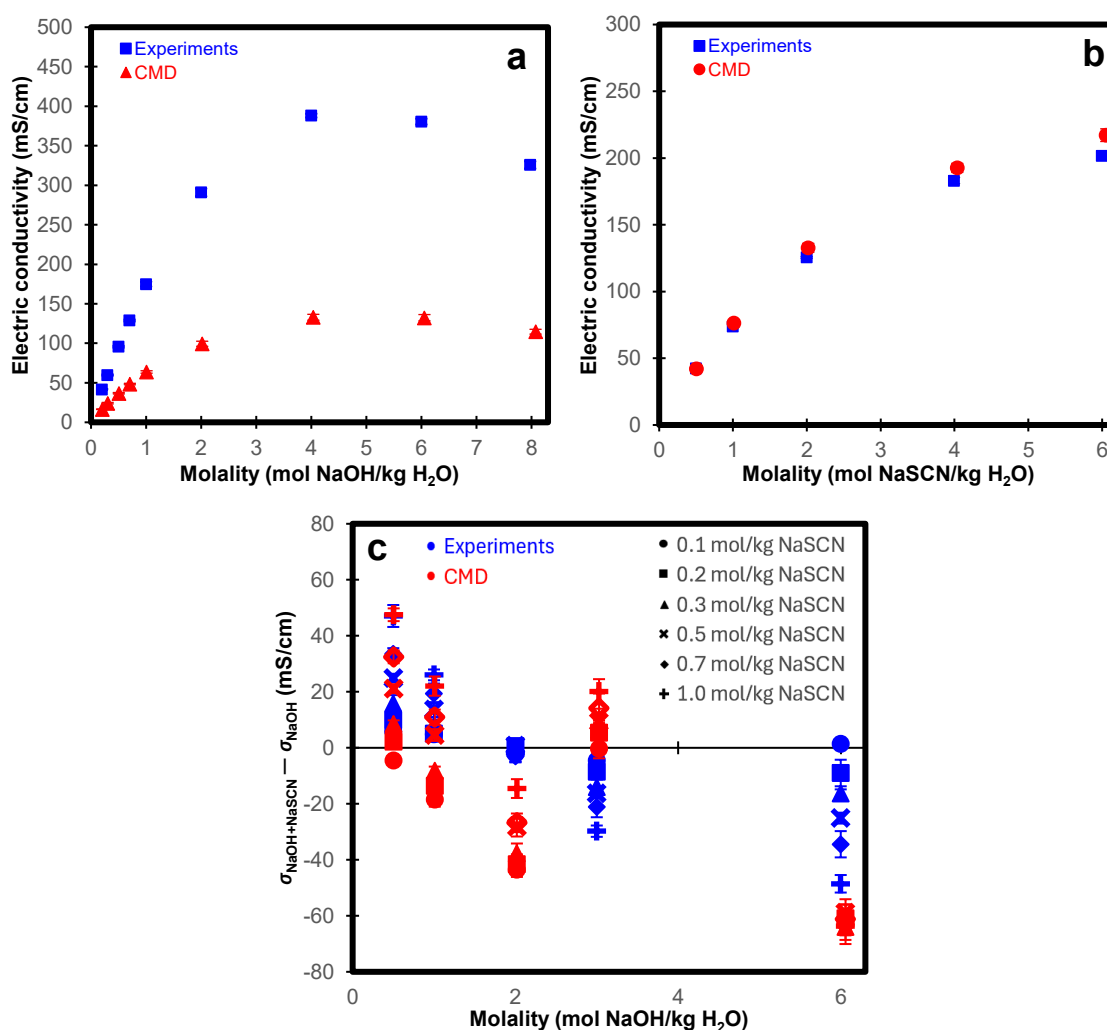


Figure 4-3 Electrical conductivity of aqueous NaOH (a), NaSCN (b) solutions as functions of concentration, and the conductivity increment resulting from the addition of NaSCN to aqueous NaOH solutions (c). The

substantial discrepancy, especially in the moderate to high NaOH concentration mixtures, between the electrical conductivities predicted by CMD simulations (red) and experimentally measured results (blue) arises primarily because CMD simulations capture only vehicular transport and do not account for the structural (Grotthuss) transport of hydroxide (OH^-) ions.

Overall, the CMD simulations successfully describe the concentration dependence of NaSCN conductivity and capture several qualitative trends in the ternary mixtures. Their systematic underestimation of pure NaOH conductivity results primarily from the omission of Grotthuss-type hydroxide transport. Given direct conductivity studies of aqueous NaOH–NaSCN mixtures appear to be limited, these results provide potentially valuable evidence of the electrolyte mixture transport behavior, but the proposed mechanism should be confirmed using reactive simulations or molecular structural transport of the hydrogen-bond network.

To estimate the contribution of structural OH^- transport, the difference between the experimentally measured conductivity and the vehicular conductivity calculated from classical MD was analyzed using the biased-random-walk model. This approach provides a corrected OH^- self-diffusion coefficient that combines the classical vehicular contribution with an estimated Grotthuss contribution. It also provides an effective OH^- lifetime associated with the time between successive proton-transfer events. Figure 4-4 presents the changes in these parameters following the addition of NaSCN into aqueous NaOH.

Equation (2-24) represents the total self-diffusion coefficient, including the structural diffusion contribution inferred from the conductivity that is not captured by classical MD simulations. A larger difference between experimental and classical conductivities therefore produces larger estimated Grotthuss contribution to the effective OH^- diffusion coefficient. The predominantly negative diffusion coefficient increments in Figure 4-4a indicate that the estimated effective OH^- mobility is lower in the NaOH–NaSCN mixtures than in the corresponding binary NaOH solutions. In term of the model, this means that adding NaSCN reduces the conductivity contribution assigned to Grotthuss transport and may also reduce the vehicular mobility calculated from the CMD trajectories. The result is consistent with NaSCN modifying the ionic and hydrogen-bond environments involved in OH^- transport. However, because the Grotthuss contribution is calculated from the conductivity difference, the diffusion results should not be presented as independent proof that SCN^- disrupt proton-transfer pathways.

The reduction in the corrected OH^- diffusion coefficient is more pronounced at low NaOH molalities partly because OH^- has a larger structural-transport contribution in the more highly hydrated solutions. Adding NaSCN can then cause a relatively large reduction in the residual conductivity $\sigma^{\text{exp}} - \sigma^{\text{C}}$. At high NaOH molality, water availability and hydroxide mobility are already restricted by the concentrated ionic environment, leaving a smaller additional structural-diffusion contribution for NaSCN to suppress.

The OH^- lifetime is also derived from the residual conductivity based on Equation (2-23), it is inversely related to the residual conductivity $\sigma^{\text{exp}} - \sigma^{\text{C}}$, although it also depends on the hopping distance, the number of Grotthuss-ions, the system volume, and the directional-correlation parameters n and q . The predominantly positive lifetime increments in Figure 4-4b therefore indicates that adding NaSCN generally decreases the conductivity assigned to proton transfer and increases the estimated time between successful OH^- hopping events. Additionally, this trend is expected because, with the increasing ion-to-water ratio, the availability of the hydrogen-bonded hydration structures required for Grotthuss proton transfer decreases. Consequently, proton hopping becomes less frequent, leading to longer transfer times. The increase in τ at higher concentrations is consistent with literature.^[64] At elevated electrolyte concentrations, other ions are more likely to occupy the first and second hydration shells of the transferring ion, disrupting the hydrogen-bond network and reducing the probability of Grotthuss transfer.

As a chaotropic and weakly hydrated anion, SCN^- can reorganize the local hydrogen-bond network of water. At an appropriate concentration, such reorganization might be expected to generate configurations favorable for OH^- proton transfer, thereby increasing the Grotthuss contribution to the effective diffusion coefficient and decreasing the estimated hopping lifetime. The present results,

however, show the opposite trend: NaSCN addition generally decreases the corrected OH^- self-diffusion coefficient and increases its estimated lifetime.

This behavior indicates that perturbation of the hydrogen-bond network does not necessarily promote structural diffusion. Efficient Grotthuss transport requires a sufficiently connected and favorably oriented sequence of hydrogen bonds; disruption of this network can reduce the continuity and directional correlation of proton-transfer pathways. Within the biased-random-walk interpretation, any potentially favorable reorientation of water induced by SCN^- appears to be outweighed by the loss of effective proton-transfer configuration. Thus, although SCN^- is conventionally described as a chaotropic ion, its net effect on OH^- transport depends on whether the resulting water structure supports connected and directionally favorable proton-transfer pathways. The present conductivity-based analysis suggests that, under the investigated conditions, NaSCN suppresses rather than enhances the Grotthuss contribution, particularly at low and moderate NaOH concentrations. The simultaneous decrease in the corrected self-diffusion coefficient and increase in the estimated lifetime are also mathematically and physically consistent. A smaller residual conductivity produces a smaller Grotthuss diffusion contribution through Equation (2-24) and, in general, a longer hopping lifetime through Equation (2-23). Nevertheless, because both quantities are derived from the experimental–CMD conductivity difference, they provide a mechanistic interpretation of the conductivity results rather than independent validation of the proposed transport mechanism, specifically in the ternary mixtures system.

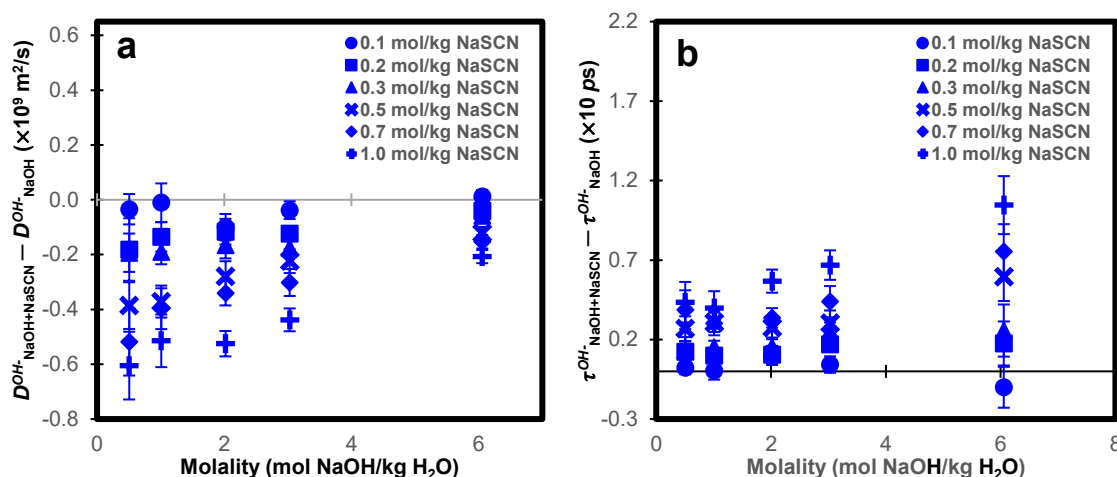


Figure 4-4 Effect of NaSCN addition on the calculated OH^- transport properties in aqueous NaOH solutions: (a) increment in the self-diffusion coefficients of OH^- and (b) increment in the estimated OH^- lifetime associated with Grotthuss transfer. The predominantly negative self-diffusion coefficient increments are accompanied by positive lifetime increments, indicating that the reduced OH^- mobility corresponds to less frequent Grotthuss transfer and, consequently, longer time between successful OH^- hopping events.

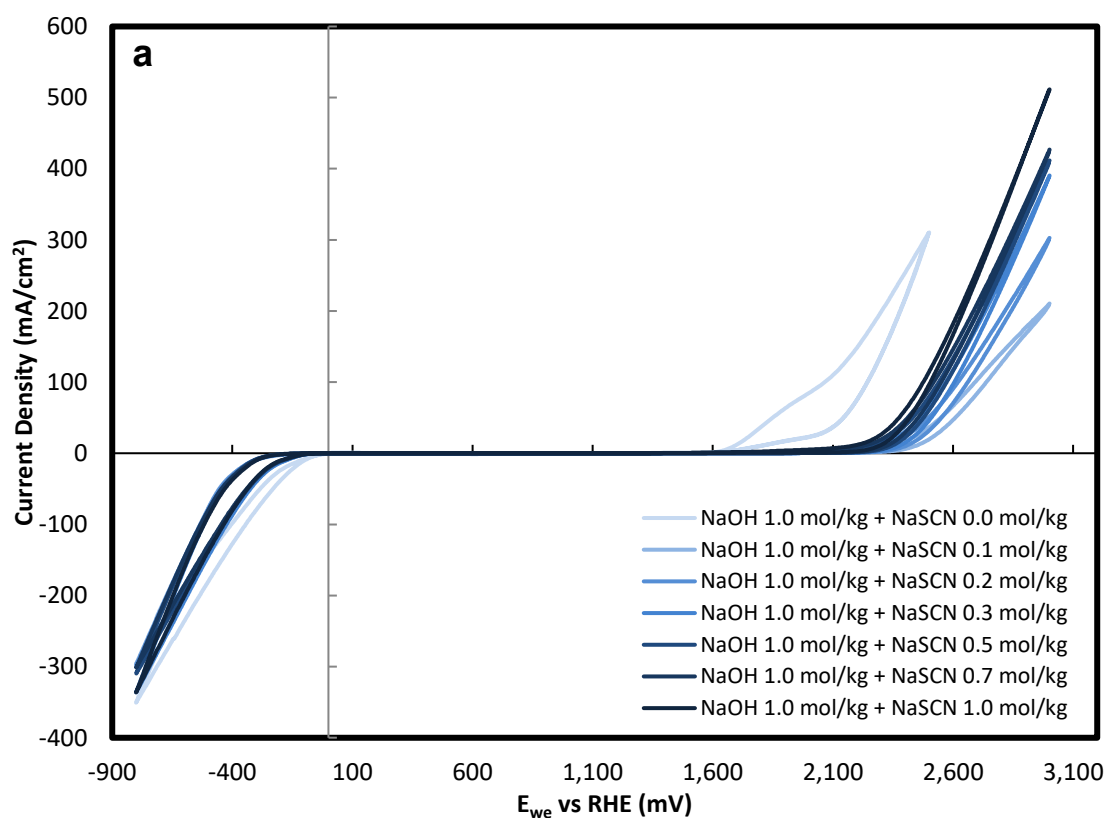
4.2 Electrochemical Analysis

Cyclic voltammetry (CV) was performed to evaluate the effect of NaSCN addition on the electrochemical performance of an NaOH electrolyte for alkaline water electrolysis. The analysis was conducted using a base electrolyte containing 1 mol/kg NaOH, with NaSCN added at concentrations ranging from 0.1 to 1 mol/kg. It is the highest NaOH concentration at which the addition of NaSCN was experimentally shown to increase the electrical conductivity of the electrolyte. Consequently, the electrochemical performance of the modified electrolyte is evaluated to determine whether the increase in electrical conductivity results in improved electrochemical activity.

Figure 4-5 shows the cyclic voltammetry polarization curve for aqueous 1 mol/kg NaOH and NaOH–NaSCN mixture obtained using a Pt-mesh electrode at 25°C, over a potential range of -0.8 V and $\sim 3.0 \text{ V}$ versus the reversible hydrogen electrode (RHE). The measurement reveals that the addition of

NaSCN shifts both the HER and OER onset away from their thermodynamic potential. The HER onset shifts from 0 V versus RHE in the NaSCN-free electrolyte to approximately -0.17 V, whereas the apparent OER onset shifts from approximately 1.6 to 2.0 V versus RHE. These shifts correspond to increased cathodic and anodic overpotentials, respectively, indicating slower apparent water-splitting kinetics following NaSCN addition.

When evaluating the electrochemical effects of introducing a chaotropic salt into an alkaline electrolyte, it is essential to consider that NaSCN increases the concentrations of Na^+ and SCN^- ions. This electrolyte engineering technique may influence electrochemical performance through both bulk and interfacial mechanisms. In the bulk electrolyte, additional ions can modify ionic conductivity and therefore the ohmic resistance associated with ionic transport^[25]. At the electrode-electrolyte interface, the ions may alter the structure of the electrical double layer, including interfacial water organization and the adsorption of reaction intermediates. These changes can affect charge-transfer kinetics and the corresponding activation overpotential.^[65]



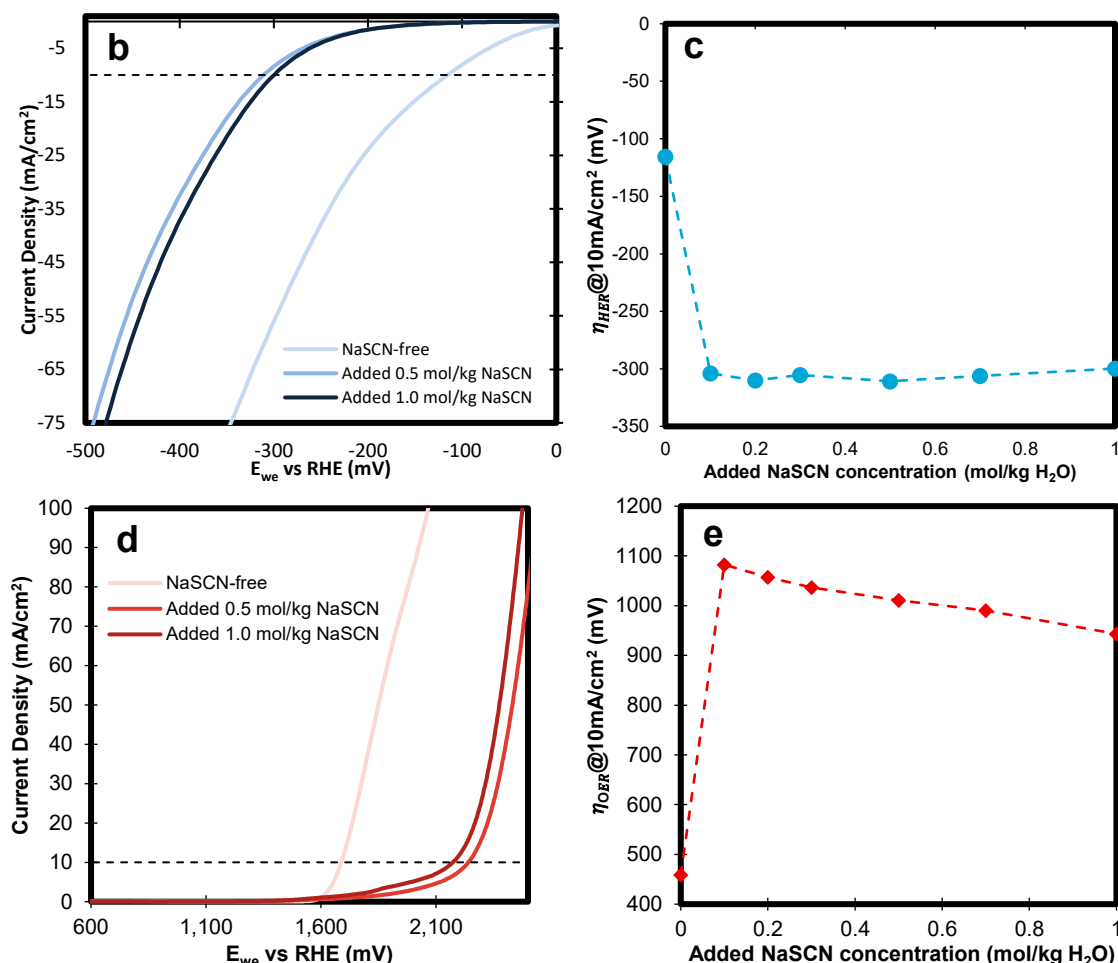


Figure 4-5 Effect of NaSCN concentration on the HER and OER polarization behavior of Pt in NaOH/NaSCN electrolytes at 25°C.

- (a) Voltammogram results of CV analysis with 1 mol NaOH / kg H₂O mixtures at 25°C.
- (b) Enlarged polarization curve of the HER potential ranges.
- (c) NaSCN concentration dependent to the required HER overpotential (η_{HER}^{10}) to achieve 10 mA/cm². Significant suppression in HER performance upon small NaSCN concentration being added is possibly attributed to the strong and rapidly saturating adsorption of SCN⁻ on Pt.
- (d) Enlarged polarization curve of the OER potential ranges.
- (e) NaSCN concentration dependent to the required OER overpotential (η_{OER}^{10}) to achieve 10 mA/cm². Significant overpotential in anodic oxidation reaction regime may be linked to competitive adsorption of SCN⁻ and OH⁻ ions on positively charged Pt anode, hindering the kinetics of OER. As the NaSCN concentration increases, the availability of SCN⁻ on the electrode also increases, promoting the activity SCN⁻ oxidation, hence the recovering slope in the graph.

The overpotentials required to reach an absolute current density of 10 mA/cm² are used to compare the apparent catalytic activities in the HER and OER region (η_{HER}^{10} and η_{OER}^{10} , respectively). A lower overpotential magnitude indicates that the target current density can be achieved with a smaller deviation from the thermodynamic equilibrium potential and therefore corresponds to better electrocatalytic performance.

The cathodic polarization curves in Figure 4-5b show that the NaSCN-free electrolyte reaches -10 mA/cm² at substantially less negative potential than the NaOH-NaSCN mixtures. The measured HER overpotential changes from -115.6 mV in 1 mol/kg NaOH to approximately -300 to -310 mV after NaSCN addition as shown in Figure 4-5c. Thus, approximately 185–195 mV of additional cathodic potential is required to reach the same current density in the presence of NaSCN, demonstrating a pronounced suppression of the HER activity.

In an AWE, the HER occurs at the negatively charge cathode. Under alkaline conditions, water is the main proton donor. The reaction begins with electrochemical dissociation of water through Volmer step as shown in Equation (2-5). The cleavage of the H–OH bond and the formation and removal of adsorbed intermediates are therefore central to alkaline HER kinetics. On Pt, the Volmer step is frequently identified as an important kinetic limitation because it requires a suitable orientation of interfacial water and simultaneous stabilization of H^* and OH^- species.^{[65], [66]}

In the electrolyte solution, water is the principal component of the electrolyte and reaches the electrode through bulk diffusion, convection, and transport through porous electrode structure. Hydrated Na^+ ions can influence the local availability, orientation, and reactivity of water within the electrical double layer (EDL). Because the Pt surface is negatively charged in the HER potential region, hydrated Na^+ ions are electrostatically attracted toward the cathode. At low concentration, the accumulation of cation can decrease the effective double-layer thickness and strengthen the interfacial electric field. The enhanced electric field promotes polarization of interfacial water molecules and may lower the activation barrier for water dissociation. At high Na^+ concentrations, however, cation–anion association becomes more pronounced, reducing the effective charge of the hydrated Na^+ and thereby causing them to move away from the negatively charged cathode surface. As a result, the EDL becomes thicker, the interfacial electrical field is reduced, water molecules become less polarized interfacial and consequently create higher activation energy for the Volmer step. This concentration dependent cation effect was demonstrated by Wang *et al.*^[67] which the study reported enhanced HER activity as the Na^+ concentrations increased from 0.001 M to ~0.1 M at constant pH, whereas further increase in Na^+ concentration ($> \sim 0.1$ M) resulted in a declined HER activity.

NaSCN may also alter the electrode–electrolyte interface. SCN^- ions accumulation or adsorption at Pt may displace interfacial water, modify the local electric field, and block sites required for the adsorption of H^* or dissociation of water. In acidic condition, SCN^- is commonly used as a Pt-site blocking probe: the addition of SCN^- has been shown to strongly suppress HER activity on Pt single-atom catalysts, demonstrating its ability to occupy active Pt sites.^[68] Although the study used a different form of Pt catalyst and electrolyte environment, it supports specific Pt–SCN interactions as a plausible explanation for the observed suppression.

Additionally, the interaction of SCN^- with Pt cannot be described solely by electrostatic attraction or repulsion. In situ FTIR measurements reported by Zhou *et al.* indicate potential-dependent specific adsorption of thiocyanate on Pt through chemical interactions with the electrode surface.^[76] Depending on the electrode potential and adsorption configuration, SCN^- may interact with Pt through either their N or S atom. Such chemisorption can compete with the adsorption of reaction intermediates and thereby inhibit catalytic reactions at the Pt surface. At anodic potentials, SCN^- accumulation near the positively polarized Pt surface may facilitate adsorption. Whereas the study suggests that at low potential, the N atom of SCN^- ion would interact and adsorbed onto Pt surface^[76], thus at cathodic potentials relevant to HER, specific Pt–NCS interactions may still enable adsorption despite unfavorable electrostatic interactions, potentially blocking sites required for hydrogen-containing intermediates and water dissociation. To distinguish among these possibilities, HER measurements should be performed at lower NaSCN concentrations to resolve the onset and saturation of inhibition, while Tafel analysis and EIS can identify changes in reaction kinetics, charge-transfer resistance, and uncompensated solution resistance.

The pronounced decrease in HER performance following the addition of a small amount of NaSCN is most plausibly attributed to the strong and rapidly saturating adsorption of SCN^- on Pt. Because SCN^- has a high affinity for Pt, even a low bulk concentration may produce near-complete coverage of the surface sites required for water dissociation and hydrogen adsorption. Adsorbed SCN^- , along with hydrated Na^+ ions, may also modify the electric environment, interfacial electric field, and hydrogen-bond structure of water near Pt, thereby affecting reaction sites beyond those occupied directly and potentially changing the HER mechanism or rate-determining step. As mentioned earlier, SCN^- has previously been used to block accessible Pt sites in catalytic studies, supporting its ability to block

accessible Pt sites in catalytic studies, supporting its ability to produce substantial activity losses at relatively low concentrations.^[68]

Based on the measured electrochemical behavior, NaSCN addition appears to introduce competing effects. Although the SCN^- ions are expected to improve HER activity by perturbing the hydrogen-bond of water network, their specific interactions to the Pt catalyst and concentration dependent cation-anion association within the double layer are shown to outweigh the force. These interactions may disrupt favorable interfacial-water configuration and blocking active Pt sites, thereby increasing the activation overpotential for alkaline HER.

The anodic polarization curves in Figure 4-5d show that NaSCN addition substantially shifts the potential required to achieve current density of 10 mA/cm^2 toward more positive values. In the NaSCN-free electrolyte, this current density is reached at 1.69 V versus RHE. Following the addition of 0.1 mol/kg NaSCN, the required potential increases sharply to 2.31 V versus RHE, indicating pronounced inhibition of the anodic activity.

For the OER, the overpotential is calculated relative to its thermodynamic equilibrium potential

$$\eta_{\text{OER}}^{10} = E_{j=10 \text{ mA/cm}^2} - 1.23 \text{ V}$$

Accordingly, the measured potential corresponds to an apparent OER overpotential of 458.6 mV in the NaSCN-free electrolyte and 1082.4 mV after adding 0.1 mol/kg NaSCN. Thus, the additional of NaSCN introduces 623.8 mV of additional anodic polarization. This substantial increase indicates that NaSCN strongly may affect the interfacial reaction kinetics and uncompensated electrolyte resistance.

The strongest inhibition is observed at the lowest investigated NaSCN addition. At concentrations above 0.1 mol/kg, the potential required to reach 10 mA/cm^2 gradually decreases from 2.31 V to 2.17 V versus RHE at 1.0 mol/kg NaSCN. Nevertheless, it remains considerably higher than that of the NaSCN-free electrolyte. The concentration dependence suggests that more than one process contributes to the anodic response. The abrupt initial increase may reflect rapid modification of the Pt surface or electrical double layer, whereas the partial recovery at higher NaSCN concentrations may involve changes in ionic conductivity, surface coverage, or additional electrochemical reactions.

Based on the material characterization of the electrolyte mixtures presented in Section 4.1, bulk transport effect may also contribute to the apparent suppression of the OER. The corrected self-diffusion coefficient of OH^- decreases following the NaSCN addition, indicating lower effective hydroxide mobility within the ternary electrolyte. Because OH^- is the principal reactant in the alkaline OER, reduced mobility can slow its transport from the bulk solution to the electrode-electrolyte interface. Under anodic polarization, this may promote a larger concentration gradient within the diffusion layer and reduce the interfacial OH^- concentration relative to the bulk value. A greater applied potential would consequently be required to maintain a given current density, contributing to the observed increase in the OER overpotential.

Similar to HER activity inhibition, specific adsorption of SCN^- provides one plausible explanation for the initial suppression.^[68] At positively polarized anode, SCN^- is electrostatically driven toward the electrode-electrolyte interface, where it may compete with OH^- for interfacial occupancy and adsorption site. Specifically, adsorbed SCN^- may occupy Pt sites required for the adsorption and conversion of oxygen-containing OER intermediates. These changes can reduce the availability of favorable configurations for interfacial electron transfer and increase the activation overpotential.

The introduction of SCN^- may also alter the potential distribution and structure of the EDL. Specific anion adsorption can modify the surface field and accessibility of the electrode to reactants. Because both SCN^- and OH^- are anions, their accumulation near the positively charged electrode may result in competitive adsorption. Preferential interfacial accumulation of SCN^- could consequently reduce the local availability of OH^- required for the OER.

Study of weakly hydrated anions such as ClO_4^- have shown that anion accumulation at electrochemical interfaces can screen water molecules and influence the activity or selectivity of water

oxidation.^{[69], [70]} By analogy, the weak hydration and high polarizability of SCN^- may favor its interfacial accumulation and enable a related screening or site-blocking effect. This comparison should nevertheless be treated cautiously because SCN^- interacts more specifically with Pt and may undergo electrochemical oxidation. Its effect may therefore involve not only double-layer restructuring but also direct adsorption, competition with OER intermediates, and parasitic thiocyanate oxidation.

Bulk and interfacial effects may act simultaneously. Reduced OH^- transport limits the replenishment of reactants near the anode, while preferential accumulation or adsorption of SCN^- may compete with OH^- for access to the positively charged surface. The resulting decrease in the local OH^- activity, combined with blockage of Pt sites and modification of interfacial-water structure, could hinder the formation and conversion of oxygen-containing intermediates. Therefore, the measured OER suppression likely reflects a combination of slower bulk ion transport, increased ohmic resistance, and less favorable interfacial reaction kinetics.

The decrease in the overpotential above 0.1 mol/kg NaSCN should not automatically be interpreted as recovery of the intrinsic OER activity. SCN^- is electrochemically active on Pt and can undergo adsorption and oxidation. Previous studies suggest that SCN^- oxidation is more readily observed under acidic system, whereas measurements using a boron-doped diamond electrode in alkaline media reported SCN^- oxidation onset at potentials above that required for OER.^{[71], [72]} However, oxidation potentials depend strongly on electrode material, surface condition, electrolyte composition and pH. Results obtained using boron-doped diamonds should be interpreted cautiously considering the utilization of Pt electrode in the present study.

Previous voltametric studies have demonstrated both adsorption and electrochemical oxidation of thiocyanate on Pt electrodes.^{[73], [74]} Consequently, some of the anodic current in the electrolytes containing NaSCN may originate from SCN^- oxidation rather than oxygen evolution. A growing contribution from SCN^- oxidation at higher concentration could cause the current density threshold to be reached at lower potential, producing the partial recovery shown in Figure 4-5e.

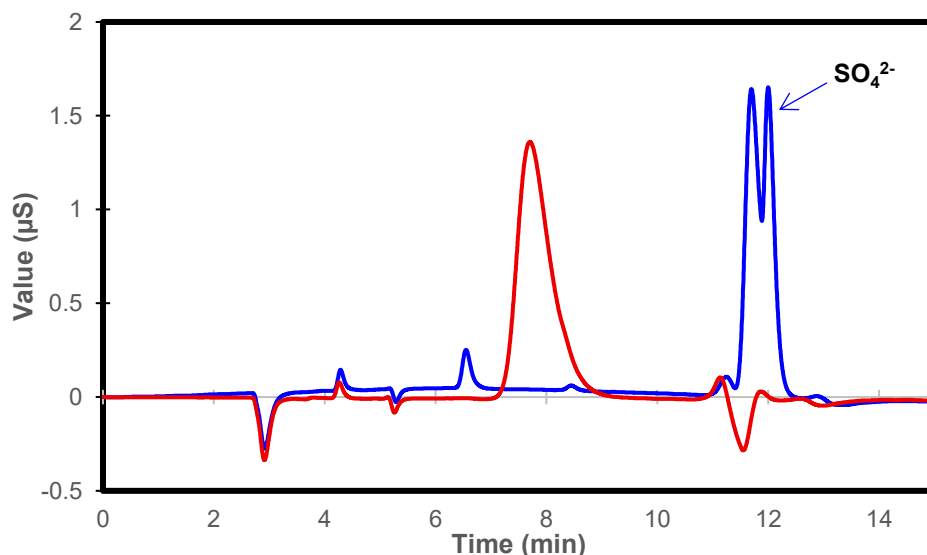
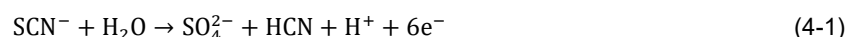


Figure 4-6 Ion chromatography analysis of the fresh NaOH/NaSCN electrolyte mixture (red) and the electrolyte collected after electrochemical experiments (blue). The ~8 min peak in the fresh electrolyte is attributed to SCN^- based on the electrolyte composition, whereas the appearance of SO_4^{2-} peak at ~12 min after electrochemical operation indicates sulfate formation and is consistent with a competing anodic oxidation pathway involving SCN^- .

Ion-chromatographic analysis of the fresh NaOH/NaSCN electrolyte shows a prominent peak at a retention time of approximately 8 minutes. Under the applied chromatographic conditions using a

Dionex IonPac AS22 column, the available manufacturer data does not provide a reference retention time that allows this peak to be clearly identified. However, considering the composition of the fresh NaOH/NaSCN mixture electrolyte, in which OH⁻ and SCN⁻ are the predominant anionic species, the peak may be attributed to SCN⁻.

Following the electrochemical measurements, a peak assigned to SO₄²⁻ appeared at a retention time of approximately 12 min.^[75] The absence of this peak in the fresh electrolyte and its appearance after electrochemical operation indicate the formation of sulfate during the experiment. This observation further provides direct evidence that SCN⁻ underwent a parasitic anodic oxidation reaction, as proposed in Equation (4-1)^[74], during the electrochemical measurement. Accordingly, the sulfur atom in SCN⁻ was therefore oxidized, partially, to sulfate, indicating that part of the anodic current is associated with a competing oxidation pathway rather than exclusively being used for oxygen evolution reaction.



The detection of sulfate supports the previous interpretation of the anodic polarization curves. In particular, the decrease in the potential required to reach 10 mA/cm² above 0.1 mol/kg NaSCN should not be regarded as an intrinsic recovery of OER activity. As the NaSCN concentration increases, the availability of SCN⁻ at the positively charged anode also increases. A progressively larger fraction of the anodic current may consequently be associated with thiocyanate oxidation. These findings indicate that the anodic current in NaSCN-containing electrolytes comprises at least two possible contributions from OER and SCN⁻ oxidation.

5 Conclusions and Recommendations

5.1 Conclusions

This study investigated the effect of sodium thiocyanate addition on the physicochemical properties and electrochemical performance of aqueous NaOH electrolytes for alkaline water electrolysis. NaSCN was selected because SCN^- is a weakly hydrated, chaotropic anion that can perturb the hydrogen-bond network of water. It was initially hypothesized that this behavior could facilitate water dissociation and thereby reduce the overpotentials associated with the hydrogen and oxygen evolution reactions. Experimental measurements were combined with empirical correlations and classical molecular dynamic simulations to evaluate this hypothesis across multiple length scales.

The material characterization results show an agreement between the experimental measurements, empirical correlation, and classical MD simulations. Based on the density and viscosity results, the MD simulations closely reproduce the experimental measurements across the investigated compositions, supporting the suitability of the parametrized force field for predicting the densities and viscosities of these electrolyte solutions. Conductivity measurements showed that NaSCN increased the number of charge carriers but did not necessarily improve ionic transport. At high NaOH and NaSCN concentrations, increased viscosity, ionic crowding, and correlated ion motion reduced mobility and produced negative conductivity increments.

Classical simulations reproduced the conductivity of aqueous NaSCN but underestimated NaOH because the nonreactive force field included vehicular transport but excluded OH^- Grotthuss transport. Within the biased-random walk model, the NaSCN addition generally decreases the corrected OH^- self-diffusion coefficient and increased the estimated hopping lifetime. These results suggest that NaSCN suppresses rather than facilitates structural OH^- transport under the investigated conditions.

Electrochemical measurements demonstrated that NaSCN substantially inhibited both the HER and apparent OER on Pt electrode. The magnitude of HER overpotential at -10 mA/cm^2 increased from 115.6 mV to 300–310 mV. Similarly, the apparent OER overpotential at 10 mA/cm^2 increased from 458.6 mV to as much as 1082.4 mV. These changes likely result from a combination of reduced OH^- mobility, increased transport resistance, modification of the EDL, and specific adsorption of SCN^- that blocks Pt sites or disrupts favorable interfacial-water configurations. Detected sulfate in the electrolyte after electrochemical measurements, indicating parasitic SCN^- oxidation. Consequently, the partial decrease in the apparent OER overpotential at higher NaSCN concentrations cannot be interpreted as recovery of intrinsic OER activity because thiocyanate oxidation may contribute to the measured anodic current density.

Overall, NaSCN did not improve alkaline water electrolysis performance under the investigated conditions. Although it increased conductivity in selected dilute mixtures, this benefit was outweighed by unfavorable bulk-transport effects, catalyst-electrolyte interactions, and limited anodic stability. These findings demonstrate that chaotropy alone is insufficient for selecting electrolyte additives. Further candidates must also maintain ionic mobility, avoid catalyst adsorption, and remain electrochemically stable throughout the operating potential range.

5.2 Recommendation for Further Research

The following items could be considered in the further research regarding the chaotropic ion effects in enhanced water electrolysis performance.

1. Future research should investigate a broader range of chaotropic and probably superchaotropic additives with weaker adsorption on selected electrocatalysts and greater electrochemical stability than SCN^- . Kosmotropic additives should be included as reference systems to clarify whether the observed effects originate specifically from chaotropicity or from more general changes in electrolyte composition. Initial screening may use readily accessible properties, including density, viscosity, conductivity, solubility, and electrochemical stability, supported by empirical correlations. Molecular dynamics simulations can subsequently be applied to the most promising candidates to examine ion hydration, hydrogen-bond network behavior, ion association, and interfacial organization. Additive selection should therefore consider not only hydrogen-bond perturbation but also ionic mobility, water activity, catalyst compatibility, and stability across the operating potential range.
2. In situ or operando Raman spectroscopy is recommended to investigate the chemical and structural behavior of the additives during electrochemical analysis. Raman measurements could identify changes in ion pairing, hydration environments, and the formation of reaction products. Complementary techniques such as infrared spectroscopy, ion chromatography, or mass spectrometry could strengthen product identification and enable more complete reaction-pathway analysis.
3. Electrochemical impedance spectroscopy and iR-corrected polarization measurements are recommended to distinguish bulk solution resistance from interfacial charge-transfer resistance. Impedance measurements over a range of additive concentrations, temperatures, and electrode potentials could determine whether performance losses originate primarily from reduced bulk conductivity, altered EDL properties, or slower reaction kinetics. Hydrogen and oxygen production should also be quantified to determine Faradaic efficiencies and separate water-splitting currents from parasitic reactions. Coupling gas product measurement with analysis of the liquid-phase products would additionally enable charge and elemental balances, providing a more reliable assessment of additive stability and reaction selectivity.

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Appendix A – Experimental Material Characterization Results

The experimental measurements of the solutions thermophysical properties—namely electrical conductivity, viscosity and density—are summarized in the following tables. All measurements were conducted at 20°C. The reported data include:

1. The molality of the solutions,
2. Mass fraction of the solutes,
3. Mass of the solvent (water) used to prepare the solutions,
4. Mass of the solutes used to prepare the solutions,
5. Results of the properties experimental measurement (3 times measurements), and
6. Average of the measured properties.

1. Experimental Electrical Conductivity Measurement
1st measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH							
20	0.200	0.008	18.18	0.1454	42.60	41.30	41.60	41.833
	0.300	0.012	10.91	0.1309	59.80	62.10	60.30	60.733
	0.500	0.020	8.37	0.1674	96.20	95.20	96.60	96.000
	0.700	0.027	12.86	0.3601	129.30	129.60	129.00	129.300
	1.000	0.038	9.26	0.3704	179.00	173.00	174.00	175.333
	2.000	0.074	7.90	0.6318	298.00	292.00	288.00	292.667
	3.001	0.107	20.44	2.4538	368.00	364.00	359.00	363.667
	4.001	0.138	7.53	1.2051	388.00	387.00	391.00	388.667
	5.996	0.193	7.98	1.9139	385.00	378.00	379.00	380.667
	7.988	0.242	8.09	2.5847	325.00	323.00	324.00	324.000

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaSCN							
20	0.500	0.039	8.00	0.3242	42.30	44.10	41.80	42.733
	1.000	0.075	8.00	0.6485	72.80	74.00	74.50	73.767
	2.001	0.140	8.00	1.2978	126.00	125.70	125.00	125.567
	4.000	0.245	8.00	2.5946	186.10	182.90	183.90	184.300
	6.001	0.327	8.00	3.8920	200.00	202.00	201.00	201.000

2nd measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH							
20	0.200	0.008	18.18	0.1454	42.5000	42.4000	41.1000	42.000
	0.300	0.012	10.91	0.1309	59.8000	59.3000	59.9000	59.667
	0.500	0.020	8.37	0.1674	95.9000	96.6000	96.4000	96.300
	0.700	0.027	12.86	0.3601	129.7000	129.1000	128.8000	129.200
	1.000	0.038	9.26	0.3704	178.4000	175.0000	175.0000	176.133
	2.000	0.074	7.90	0.6318	297.0000	286.0000	290.0000	291.000
	3.001	0.107	20.44	2.45	359.00	362.00	362.00	361.000
	4.001	0.138	7.53	1.2051	387.0000	387.0000	392.0000	388.667
	5.996	0.193	7.98	1.9139	374.0000	377.0000	381.0000	377.333
	7.988	0.242	8.09	2.5847	330.0000	324.0000	318.0000	324.000

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaSCN							
20	0.500	0.039	8.00	0.3242	41.80	43.10	42.60	42.500
	1.000	0.075	8.00	0.6485	72.80	73.60	74.40	73.600
	2.001	0.140	8.00	1.2978	125.90	125.40	126.00	125.767
	4.000	0.245	8.00	2.5946	184.30	180.60	183.60	182.833
	6.001	0.327	8.00	3.8920	200.00	204.00	203.00	202.333

3rd measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH							
20	0.200	0.008	18.18	0.1454	41.90	40.50	40.90	41.100
	0.300	0.012	10.91	0.1309	58.90	59.40	59.10	59.133
	0.500	0.020	8.37	0.1674	95.50	92.70	95.30	94.500
	0.700	0.027	12.86	0.3601	130.10	125.80	128.10	128.000
	1.000	0.038	9.26	0.3704	177.10	169.50	172.00	172.867
	2.000	0.074	7.90	0.6318	295.00	283.00	290.00	289.333
	3.001	0.107	20.44	2.45	361.00	363.00	360.00	361.333
	4.001	0.138	7.53	1.2051	390.00	384.00	387.00	387.000
	5.996	0.193	7.98	1.9139	386.00	383.00	382.00	383.667
	7.988	0.242	8.09	2.5847	333.00	332.00	323.00	329.333

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaSCN							
20	0.500	0.039	8.00	0.3242	41.70	42.40	41.90	42.000
	1.000	0.075	8.00	0.6485	73.00	73.90	74.30	73.733
	2.001	0.140	8.00	1.2978	126.00	124.00	126.10	125.367
	4.000	0.245	8.00	2.5946	184.20	177.70	183.70	181.867
	6.001	0.327	8.00	3.8920	203.00	199.90	202.00	201.633

1st measurements

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN					
Mixed Solution											
20	0.500	0.000	0.0196	0.0000	0.1567	0.0000	7.8433	96.20	95.9000	95.50	95.86667
	0.500	0.100	0.0194	0.0079	0.1567	0.0639	7.8433	102.30	101.60	99.10	101.00000
	0.500	0.201	0.0193	0.0157	0.1567	0.1275	7.8433	106.70	105.70	104.90	105.76667
	0.500	0.302	0.0191	0.0234	0.1567	0.1919	7.8433	112.00	113.10	108.30	111.13333
	0.500	0.500	0.0188	0.0382	0.1567	0.3179	7.8433	122.30	120.30	119.70	120.76667
	0.500	0.700	0.0186	0.0527	0.1567	0.4450	7.8433	130.10	129.90	127.70	129.23333
	0.500	1.000	0.0181	0.0736	0.1567	0.6356	7.8433	144.90	141.70	141.20	142.60000
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	179.00	178.40	177.10	178.17
	1.000	0.100	0.0382	0.0078	0.3077	0.0626	7.6923	182.70	181.10	179.00	180.93
	1.000	0.200	0.0379	0.0154	0.3077	0.1249	7.6923	179.60	179.80	176.30	178.57
	1.000	0.301	0.0376	0.0230	0.3077	0.1880	7.6923	182.10	181.60	181.10	181.60
	1.000	0.500	0.0370	0.0375	0.3077	0.3118	7.6923	188.80	187.30	187.50	187.87
	1.000	0.700	0.0365	0.0518	0.3077	0.4367	7.6923	195.40	193.30	194.30	194.33
	1.000	1.000	0.0357	0.0723	0.3077	0.6236	7.6923	199.80	201.00	199.30	200.03
20	2.001	0.000	0.0741	0.0000	0.5927	0.0000	7.4073	298.00	297.00	295.00	296.67
	2.001	0.102	0.0735	0.0076	0.5927	0.0610	7.4073	288.00	285.00	284.00	285.67
	2.001	0.201	0.0730	0.0149	0.5927	0.1206	7.4073	286.00	290.00	290.00	288.67
	2.001	0.301	0.0725	0.0221	0.5927	0.1805	7.4073	291.00	292.00	286.00	289.67
	2.001	0.501	0.0714	0.0362	0.5927	0.3008	7.4073	294.00	295.00	291.00	293.33
	2.001	0.700	0.0704	0.0499	0.5927	0.4204	7.4073	290.00	286.00	285.00	287.00
	2.001	1.000	0.0689	0.0698	0.5927	0.6005	7.4073	288.00	286.00	283.00	285.67
20	3.001	0.000	0.1072	0.0000	2.4538	0.0000	20.4429	368.00	364.00	359.00	363.67
	3.000	0.101	0.1064	0.0072	0.8571	0.0583	7.1429	357.00	354.00	356.00	355.67

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN					
	3.000	0.200	0.1056	0.0143	0.8571	0.1160	7.1429	353.00	352.00	353.00	352.67
	3.001	0.301	0.1049	0.0213	0.8573	0.1743	7.1427	345.00	346.00	349.00	346.67
	3.000	0.500	0.1034	0.0349	0.8571	0.2897	7.1429	348.00	346.00	347.00	347.00
	3.000	0.700	0.1020	0.0483	0.8571	0.4056	7.1429	345.00	339.00	343.00	342.33
	3.000	1.001	0.0999	0.0676	0.8571	0.5796	7.1429	337.00	330.00	331.00	332.67
20	4.0002	0.0000	0.1379	0.0000	7.6868	0.0000	48.0400	390.00	387.00	393.00	390.00
	4.0002	0.1002	0.1370	0.0070	1.1035	0.0560	6.8965	385.00	385.00	386.00	385.33
	4.0002	0.2000	0.1360	0.0138	1.1035	0.1118	6.8965	381.00	382.00	379.00	380.67
	4.0002	0.2999	0.1351	0.0205	1.1035	0.1677	6.8965	376.00	377.00	376.00	376.33
	4.0002	0.4999	0.1333	0.0338	1.1035	0.2795	6.8965	372.00	370.00	370.00	370.67
	4.0002	0.7001	0.1315	0.0466	1.1035	0.3914	6.8965	362.00	362.00	364.00	362.67
	4.0002	1.0007	0.1289	0.0654	1.1035	0.5595	6.8965	352.00	353.00	354.00	353.00
20	6.00	0.00	0.19	0.00	1.55	0.00	6.45	385.00	374.00	386.00	381.67
	6.00	0.10	0.19	0.01	1.55	0.05	6.45	388.00	382.00	388.00	386.00
	6.00	0.20	0.19	0.01	1.55	0.10	6.45	371.00	374.00	378.00	374.33
	6.00	0.30	0.19	0.02	1.55	0.16	6.45	364.00	363.00	366.00	364.33
	6.00	0.50	0.19	0.03	1.55	0.26	6.45	356.00	358.00	358.00	357.33
	6.00	0.70	0.19	0.04	1.55	0.37	6.45	343.00	347.00	350.00	346.67
	6.00	1.00	0.18	0.06	1.55	0.52	6.45	332.00	332.00	339.00	334.33

2nd measurements

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN					
Mixed Solution											
20	0.500	0.000	0.0196	0.0000	0.1567	0.0000	7.8433	95.2000	96.60	92.70	94.83333
	0.500	0.100	0.0194	0.0079	0.1567	0.0639	7.8433	103.6000	102.00	101.40	102.33333
	0.500	0.201	0.0193	0.0157	0.1567	0.1275	7.8433	107.0000	105.80	105.00	105.93333
	0.500	0.302	0.0191	0.0234	0.1567	0.1919	7.8433	113.8000	111.20	110.10	111.70000
	0.500	0.500	0.0188	0.0382	0.1567	0.3179	7.8433	122.1000	120.00	118.60	120.23333
	0.500	0.700	0.0186	0.0527	0.1567	0.4450	7.8433	130.5000	129.20	127.40	129.03333
	0.500	1.000	0.0181	0.0736	0.1567	0.6356	7.8433	145.6000	141.50	141.10	142.73333
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	173.0000	175.00	169.50	172.5000
	1.000	0.100	0.0382	0.0078	0.3077	0.0626	7.6923	178.2000	179.30	179.00	178.8333
	1.000	0.200	0.0379	0.0154	0.3077	0.1249	7.6923	181.2000	180.20	180.20	180.5333
	1.000	0.301	0.0376	0.0230	0.3077	0.1880	7.6923	184.4000	181.00	182.70	182.7000
	1.000	0.500	0.0370	0.0375	0.3077	0.3118	7.6923	189.1000	186.80	188.40	188.1000
	1.000	0.700	0.0365	0.0518	0.3077	0.4367	7.6923	194.3000	192.70	196.50	194.5000
	1.000	1.000	0.0357	0.0723	0.3077	0.6236	7.6923	202.0000	200.00	199.30	200.4333
20	2.001	0.000	0.0741	0.0000	0.5927	0.0000	7.4073	292.0000	286.00	283.00	287.00
	2.001	0.102	0.0735	0.0076	0.5927	0.0610	7.4073	292.0000	288.00	287.00	289.00
	2.001	0.201	0.0730	0.0149	0.5927	0.1206	7.4073	294.0000	288.00	288.00	290.00
	2.001	0.301	0.0725	0.0221	0.5927	0.1805	7.4073	295.0000	289.00	286.00	290.00
	2.001	0.501	0.0714	0.0362	0.5927	0.3008	7.4073	289.0000	289.00	286.00	288.00
	2.001	0.700	0.0704	0.0499	0.5927	0.4204	7.4073	287.0000	288.00	285.00	286.67
	2.001	1.000	0.0689	0.0698	0.5927	0.6005	7.4073	284.0000	287.00	284.00	285.00
20	3.001	0.000	0.1072	0.0000	2.4538	0.0000	20.4429	359.0000	362.00	362.00	361.00

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN					
	3.000	0.101	0.1064	0.0072	0.8571	0.0583	7.1429	362.0000	358.00	357.00	359.00
	3.000	0.200	0.1056	0.0143	0.8571	0.1160	7.1429	355.0000	355.00	356.00	355.33
	3.001	0.301	0.1049	0.0213	0.8573	0.1743	7.1427	352.0000	349.00	347.00	349.33
	3.000	0.500	0.1034	0.0349	0.8571	0.2897	7.1429	350.0000	343.00	345.00	346.00
	3.000	0.700	0.1020	0.0483	0.8571	0.4056	7.1429	347.0000	337.00	340.00	341.33
	3.000	1.001	0.0999	0.0676	0.8571	0.5796	7.1429	334.0000	331.00	332.00	332.33
20	3.9997	0.0000	0.1379	0.0000	7.6442	0.0000	47.78	392.00	388.00	387.00	389.00
	3.9997	0.1003	0.1370	0.0070	1.1034	0.0561	6.8966	385.00	388.00	384.00	385.67
	3.9997	0.2005	0.1360	0.0138	1.1034	0.1121	6.8966	380.00	381.00	382.00	381.00
	3.9997	0.2999	0.1351	0.0205	1.1034	0.1677	6.8966	377.00	375.00	377.00	376.33
	3.9997	0.5006	0.1333	0.0338	1.1034	0.2799	6.8966	370.00	370.00	371.00	370.33
	3.9997	0.7000	0.1315	0.0466	1.1034	0.3914	6.8966	362.00	362.00	362.00	362.00
	3.9997	0.9998	0.1289	0.0653	1.1034	0.5590	6.8966	351.00	352.00	350.00	351.00
20	6.00	0.00	0.19	0.00	1.55	0.00	6.45	378.00	377.00	383.00	379.33
	6.00	0.10	0.19	0.01	1.55	0.05	6.45	388.00	381.00	376.00	381.67
	6.00	0.20	0.19	0.01	1.55	0.10	6.45	364.00	374.00	370.00	369.33
	6.00	0.30	0.19	0.02	1.55	0.16	6.45	365.00	367.00	360.00	364.00
	6.00	0.50	0.19	0.03	1.55	0.26	6.45	353.00	358.00	355.00	355.33
	6.00	0.70	0.19	0.04	1.55	0.37	6.45	348.00	346.00	347.00	347.00
	6.00	1.00	0.18	0.06	1.55	0.52	6.45	329.00	332.00	331.00	330.67

3rd measurements

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN					
Mixed Solution											
20	0.500	0.000	0.0196	0.0000	0.1567	0.0000	7.8433	96.6000	96.4000	95.30	96.10000
	0.500	0.100	0.0194	0.0079	0.1567	0.0639	7.8433	101.7000	101.50	100.20	101.13333
	0.500	0.201	0.0193	0.0157	0.1567	0.1275	7.8433	104.9000	105.00	104.40	104.76667
	0.500	0.302	0.0191	0.0234	0.1567	0.1919	7.8433	112.2000	110.90	110.50	111.20000
	0.500	0.500	0.0188	0.0382	0.1567	0.3179	7.8433	121.3000	120.60	119.50	120.46667
	0.500	0.700	0.0186	0.0527	0.1567	0.4450	7.8433	130.3000	128.40	128.30	129.00000
	0.500	1.000	0.0181	0.0736	0.1567	0.6356	7.8433	143.0000	142.60	142.50	142.70000
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	174.0000	175.00	172.00	173.6667
	1.000	0.100	0.0382	0.0078	0.3077	0.0626	7.6923	179.3000	180.70	177.50	179.1667
	1.000	0.200	0.0379	0.0154	0.3077	0.1249	7.6923	180.8000	181.40	178.70	180.3000
	1.000	0.301	0.0376	0.0230	0.3077	0.1880	7.6923	183.6000	182.60	183.40	183.2000
	1.000	0.500	0.0370	0.0375	0.3077	0.3118	7.6923	189.0000	191.40	187.50	189.3000
	1.000	0.700	0.0365	0.0518	0.3077	0.4367	7.6923	193.7000	193.20	193.90	193.6000
	1.000	1.000	0.0357	0.0723	0.3077	0.6236	7.6923	203.0000	202.00	201.00	202.0000
20	2.001	0.000	0.0741	0.0000	0.5927	0.0000	7.4073	288.0000	290.00	290.00	289.33
	2.001	0.102	0.0735	0.0076	0.5927	0.0610	7.4073	292.0000	293.00	291.00	292.00
	2.001	0.201	0.0730	0.0149	0.5927	0.1206	7.4073	292.0000	290.00	287.00	289.67
	2.001	0.301	0.0725	0.0221	0.5927	0.1805	7.4073	288.0000	289.00	288.00	288.33
	2.001	0.501	0.0714	0.0362	0.5927	0.3008	7.4073	290.0000	288.00	289.00	289.00
	2.001	0.700	0.0704	0.0499	0.5927	0.4204	7.4073	287.0000	286.00	286.00	286.33
	2.001	1.000	0.0689	0.0698	0.5927	0.6005	7.4073	285.0000	284.00	286.00	285.00
20	3.001	0.000	0.1072	0.0000	2.4538	0.0000	20.4429	361.0000	363.00	360.00	361.33

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	Solution exp conductivity / mS/cm	AVERAGE Solution exp conductivity / mS/cm
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN					
	3.000	0.101	0.1064	0.0072	0.8571	0.0583	7.1429	358.0000	357.00	358.00	357.67
	3.000	0.200	0.1056	0.0143	0.8571	0.1160	7.1429	352.0000	355.00	350.00	352.33
	3.001	0.301	0.1049	0.0213	0.8573	0.1743	7.1427	349.0000	348.00	347.00	348.00
	3.000	0.500	0.1034	0.0349	0.8571	0.2897	7.1429	344.0000	345.00	344.00	344.33
	3.000	0.700	0.1020	0.0483	0.8571	0.4056	7.1429	336.0000	338.00	343.00	339.00
	3.000	1.001	0.0999	0.0676	0.8571	0.5796	7.1429	333.0000	332.00	330.00	331.67
20	4.00	0.00	0.14	0.00	7.6442	0.0000	47.78	392.00	391.00	390.00	391.00
	4.00	0.10	0.14	0.01	1.1034	0.0561	6.8966	386.00	385.00	385.00	385.33
	4.00	0.20	0.14	0.01	1.1034	0.1121	6.8966	380.00	379.00	379.00	379.33
	4.00	0.30	0.14	0.02	1.1034	0.1677	6.8966	376.00	377.00	378.00	377.00
	4.00	0.50	0.13	0.03	1.1034	0.2799	6.8966	369.00	368.00	368.00	368.33
	4.00	0.70	0.13	0.05	1.1034	0.3914	6.8966	364.00	362.00	361.00	362.33
	4.00	1.00	0.13	0.07	1.1034	0.5590	6.8966	355.00	353.00	353.00	353.67
20	6.00	0.00	0.19	0.00	1.55	0.00	6.45	379.00	381.00	382.00	380.67
	6.00	0.10	0.19	0.01	1.55	0.05	6.45	376.00	381.00	378.00	378.33
	6.00	0.20	0.19	0.01	1.55	0.10	6.45	369.00	369.00	375.00	371.00
	6.00	0.30	0.19	0.02	1.55	0.16	6.45	365.00	365.00	363.00	364.33
	6.00	0.50	0.19	0.03	1.55	0.26	6.45	349.00	359.00	353.00	353.67
	6.00	0.70	0.19	0.04	1.55	0.37	6.45	344.00	345.00	345.00	344.67
	6.00	1.00	0.18	0.06	1.55	0.52	6.45	328.00	335.00	330.00	331.00

2. Experimental Viscosity Measurement

1st measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp viscosity / mPa s
	NaOH				
20	0.200	0.008	18.18	0.1454	1.032
	0.300	0.012	10.91	0.1309	1.046
	0.500	0.020	7.84	0.1567	1.091
	0.700	0.027	12.86	0.3601	1.154
	1.000	0.038	32.34	1.2940	1.200
	2.001	0.074	7.41	0.5927	1.509
	4.001	0.138	7.53	1.2051	2.540
	6.000	0.194	31.51	7.5616	4.248
	7.988	0.242	8.09	2.5847	7.140

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp viscosity / mPa s
	NaSCN				
20	0.500	0.039	8.00	0.3242	1.0071
	1.000	0.075	8.00	0.6485	1.0348
	2.001	0.140	8.00	1.2978	1.1337
	4.000	0.245	8.00	2.5946	1.4034
	6.001	0.327	8.00	3.8920	1.8195

2nd measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp viscosity / mPa s
	NaOH				
20	0.200	0.008	18.18	0.1454	1.0210
	0.300	0.012	10.91	0.1309	1.0637
	0.500	0.020	7.8431	0.1569	1.1066
	0.700	0.027	12.86	0.3601	1.1712
	1.000	0.038	7.69	0.3077	1.2442
	2.000	0.074	7.41	0.5925	1.5009
	4.001	0.138	7.53	1.2051	2.5359
	6.001	0.194	6.45	1.5483	4.2731
	7.988	0.242	8.09	2.5847	7.2425

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp viscosity / mPa s
	NaSCN				
20	0.500	0.039	6.01	0.2438	1.0097
	1.001	0.075	6.00	0.4869	1.0370
	1.994	0.139	6.02	0.9733	1.1271
	3.992	0.245	6.01	1.9450	1.4130
	5.993	0.327	8.01	3.8919	1.8222

3rd measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp viscosity / mPa s
	NaOH				
20	0.200	0.008	18.18	0.1454	1.0368
	0.300	0.012	10.91	0.1309	1.0557
	0.500	0.020	7.8430	0.1570	1.1042
	0.700	0.027	12.86	0.3601	1.1621
	1.000	0.038	7.69	0.3077	1.2433
	2.000	0.074	7.41	0.5925	1.5167
	4.001	0.138	7.53	1.2051	2.5346
	6.001	0.194	6.45	1.5483	4.3274
	7.988	0.242	8.09	2.5847	7.2400

T /°C	Molality / mol/kg	Mass Fraction	Water Mass / g	Solute Mass / gr	Solution exp viscosity / mPa s
	NaSCN				
20	0.501	0.039	8.03	0.3260	0.9978
	0.999	0.075	8.02	0.6496	1.0396
	1.998	0.139	8.01	1.2977	1.1487
	3.988	0.244	8.03	2.5959	1.4068
	6.001	0.327	8.00	3.8922	1.8156

1st measurements

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp viscosity / mPa s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN		
Mixed Solution								
20	0.500	0.000	0.0196	0.0000	0.1567	0.0000	7.8433	1.0908
	0.500	0.100	0.0194	0.0079	0.1567	0.0639	7.8433	1.0628
	0.500	0.201	0.0193	0.0157	0.1567	0.1275	7.8433	1.0637
	0.500	0.302	0.0191	0.0234	0.1567	0.1919	7.8433	1.0946
	0.500	0.500	0.0188	0.0382	0.1567	0.3179	7.8433	1.1019
	0.500	0.700	0.0186	0.0527	0.1567	0.4450	7.8433	1.1535
	0.500	1.000	0.0181	0.0736	0.1567	0.6356	7.8433	1.1902
20	1.000	0.000	0.0385	0.0000	1.2940	0.0000	32.3406	1.2003
	1.000	0.100	0.0382	0.0077	0.2412	0.0489	6.0300	1.2427
	1.000	0.201	0.0379	0.0154	0.2308	0.0940	5.7692	1.2451
	1.000	0.301	0.0376	0.0230	0.3077	0.1880	7.6923	1.2691
	1.000	0.500	0.0370	0.0375	0.3077	0.3118	7.6923	1.2823
	1.000	0.700	0.0365	0.0518	0.3077	0.4367	7.6923	1.3213
	1.000	1.000	0.0357	0.0723	0.2319	0.4700	5.7981	1.3385
20	2.001	0.000	0.0741	0.0000	0.5927	0.0000	7.4073	1.5094
	2.001	0.102	0.0735	0.0076	0.5927	0.0610	7.4073	1.5111
	2.001	0.201	0.0730	0.0149	0.5927	0.1206	7.4073	1.5860
	2.001	0.301	0.0725	0.0221	0.5927	0.1805	7.4073	1.6065
	2.001	0.501	0.0714	0.0362	0.5927	0.3008	7.4073	1.6198
	2.001	0.700	0.0704	0.0499	0.5927	0.4204	7.4073	1.6638
	2.001	1.000	0.0689	0.0698	0.5927	0.6005	7.4073	1.7469
20	2.997	0.000	0.1070	0.0000	0.9745	0.0000	8.1301	1.9934

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp viscosity / mPa s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN		
	3.000	0.100	0.1064	0.0072	0.6923	0.0470	5.7692	2.0298
	3.000	0.199	0.1056	0.0142	0.6923	0.0932	5.7692	2.0061
	3.001	0.301	0.1049	0.0213	0.8573	0.1743	7.1427	1.9927
	3.000	0.500	0.1034	0.0349	0.8571	0.2897	7.1429	2.0707
	3.000	0.700	0.1020	0.0483	0.8571	0.4056	7.1429	2.1767
	3.000	0.999	0.0999	0.0674	0.6923	0.4673	5.7692	2.2629
20	6.000	0.000	0.1935	0.0000	7.5616	0.0000	31.5070	4.2482
	6.000	0.101	0.1923	0.0065	1.1613	0.0394	4.8387	4.2365
	6.000	0.200	0.1911	0.0129	1.1613	0.0783	4.8387	4.4608
	6.001	0.300	0.1899	0.0192	1.5486	0.1567	6.4514	4.4911
	6.001	0.500	0.1874	0.0317	1.5486	0.2616	6.4514	4.5734
	6.001	0.701	0.1851	0.0438	1.5486	0.3667	6.4514	4.6786
	6.000	0.998	0.1817	0.0612	1.1625	0.3918	4.8440	4.7988

2nd measurements

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp viscosity / mPa s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN		
Mixed Solution								
20	0.500	0.000	0.0196	0.0000	0.1569	0.0000	7.8431	1.1066
	0.500	0.101	0.0195	0.0079	0.1569	0.0641	7.8431	1.0350
	0.500	0.201	0.0193	0.0157	0.1569	0.1277	7.8431	1.0431
	0.500	0.300	0.0192	0.0233	0.1569	0.1909	7.8431	1.0774
	0.500	0.500	0.0189	0.0382	0.1569	0.3179	7.8431	1.0862

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp viscosity / mPa s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN		
	0.500	0.700	0.0186	0.0527	0.1569	0.4453	7.8431	1.1808
	0.500	1.001	0.0182	0.0737	0.1569	0.6367	7.8431	1.1837
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	1.2442
	1.001	0.100	0.0382	0.0078	0.3079	0.0625	7.6921	1.2358
	1.001	0.200	0.0379	0.0154	0.3079	0.1249	7.6921	1.2086
	1.001	0.301	0.0376	0.0229	0.3079	0.1875	7.6921	1.2649
	1.001	0.501	0.0370	0.0376	0.3079	0.3124	7.6921	1.2864
	1.001	0.701	0.0365	0.0518	0.3079	0.4371	7.6921	1.3155
	1.001	1.000	0.0357	0.0723	0.3079	0.6238	7.6921	1.3258
20	2.000	0.000	0.0741	0.0000	0.5925	0.0000	7.4075	1.5009
	2.000	0.100	0.0735	0.0075	0.5925	0.0602	7.4075	1.5410
	2.000	0.200	0.0730	0.0148	0.5925	0.1204	7.4075	1.5735
	2.000	0.300	0.0724	0.0220	0.5925	0.1803	7.4075	1.6176
	2.000	0.501	0.0714	0.0362	0.5925	0.3006	7.4075	1.6097
	2.000	0.701	0.0704	0.0500	0.5925	0.4208	7.4075	1.6622
	2.000	1.000	0.0689	0.0699	0.5925	0.6008	7.4075	1.7469
20	3.000	0.000	0.1071	0.0000	0.8571	0.0000	7.1429	1.9873
	3.000	0.100	0.1064	0.0072	0.8571	0.0580	7.1429	1.9671
	3.000	0.200	0.1056	0.0142	0.8571	0.1156	7.1429	2.0060
	3.000	0.302	0.1049	0.0214	0.8571	0.1746	7.1429	2.0292
	3.000	0.500	0.1034	0.0350	0.8571	0.2898	7.1429	2.0501
	3.000	0.701	0.1020	0.0483	0.8571	0.4059	7.1429	2.1725
	3.000	1.000	0.0999	0.0675	0.8571	0.5791	7.1429	2.1864
20	6.000	0.000	0.1936	0.0000	1.5483	0.0000	6.4510	4.2731

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp viscosity / mPa s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN		
	6.000	0.100	0.1923	0.0065	1.5483	0.0523	6.4510	4.3448
	6.001	0.201	0.1911	0.0130	1.5486	0.1053	6.4514	4.4323
	6.000	0.300	0.1898	0.0192	1.5484	0.1569	6.4516	4.4915
	6.000	0.501	0.1874	0.0317	1.5484	0.2619	6.4516	4.6382
	6.000	0.700	0.1851	0.0438	1.5484	0.3663	6.4516	4.6847
	6.001	1.000	0.1817	0.0614	1.5486	0.5232	6.4514	4.8523

3rd measurements

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp viscosity / mPa s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN		
Mixed Solution								
20	0.500	0.000	0.0196	0.0000	0.1570	0.0000	7.8430	1.1042
	0.500	0.101	0.0195	0.0079	0.1570	0.0640	7.8430	1.0368
	0.500	0.200	0.0193	0.0157	0.1570	0.1272	7.8430	1.0280
	0.500	0.300	0.0192	0.0233	0.1570	0.1909	7.8430	1.0940
	0.500	0.500	0.0189	0.0382	0.1570	0.3180	7.8430	1.1119
	0.500	0.700	0.0186	0.0527	0.1570	0.4453	7.8430	1.1748
	0.500	1.000	0.0182	0.0737	0.1570	0.6361	7.8430	1.1788
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	1.2433
	1.001	0.100	0.0382	0.0077	0.3079	0.0623	7.6921	1.2433
	1.001	0.200	0.0379	0.0154	0.3079	0.1248	7.6921	1.2495
	1.001	0.300	0.0376	0.0229	0.3079	0.1871	7.6921	1.2550
	1.001	0.501	0.0370	0.0376	0.3079	0.3123	7.6921	1.2645

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution exp viscosity / mPa s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN		
	1.001	0.700	0.0365	0.0518	0.3079	0.4368	7.6921	1.3117
	1.001	1.000	0.0357	0.0723	0.3079	0.6238	7.6921	1.3223
20	2.000	0.000	0.0741	0.0000	0.5925	0.0000	7.4075	1.5167
	2.000	0.100	0.0735	0.0075	0.5925	0.0603	7.4075	1.5303
	2.000	0.200	0.0730	0.0148	0.5925	0.1201	7.4075	1.5716
	2.000	0.301	0.0724	0.0221	0.5925	0.1808	7.4075	1.6094
	2.000	0.501	0.0714	0.0362	0.5925	0.3006	7.4075	1.6253
	2.000	0.701	0.0704	0.0500	0.5925	0.4208	7.4075	1.6683
	2.000	1.000	0.0689	0.0698	0.5925	0.6005	7.4075	1.7447
20	3.000	0.000	0.1071	0.0000	0.8571	0.0000	7.1429	1.9741
	3.000	0.101	0.1064	0.0072	0.8571	0.0583	7.1429	2.0476
	3.000	0.201	0.1056	0.0143	0.8571	0.1164	7.1429	2.0355
	3.000	0.300	0.1049	0.0212	0.8571	0.1735	7.1429	2.0300
	3.000	0.500	0.1034	0.0349	0.8571	0.2897	7.1429	2.0855
	3.000	0.701	0.1020	0.0483	0.8571	0.4058	7.1429	2.1702
	3.000	1.001	0.0999	0.0676	0.8571	0.5798	7.1429	2.1999
20	6.000	0.000	0.1936	0.0000	1.5483	0.0000	6.4510	4.3274
	6.000	0.101	0.1923	0.0066	1.5483	0.0529	6.4510	4.4353
	6.001	0.201	0.1911	0.0130	1.5486	0.1053	6.4514	4.4725
	6.001	0.300	0.1898	0.0193	1.5486	0.1571	6.4514	4.6091
	6.001	0.500	0.1875	0.0317	1.5486	0.2615	6.4514	4.6618
	6.001	0.701	0.1851	0.0438	1.5486	0.3664	6.4514	4.6986
	6.001	1.000	0.1817	0.0614	1.5486	0.5230	6.4514	4.8539

3. Experimental Density Measurement

1st measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Volume / mL	Solute Mass / gr	Density from literature / kg/m³	Density ^{exp} / kg/m³	Density Residual / kg/m³
	NaOH						
20	0.000	0.000	0.00	0.00	998.20	998.20	0.000
	0.200	0.008	18.18	0.1454	1006.88	1007.09	0.214
	0.300	0.012	10.91	0.1309	1011.17	1011.37	0.201
	0.500	0.020	8.37	0.1674	1019.67	1019.88	0.210
	0.700	0.027	12.86	0.3601	1028.06	1028.34	0.284
	1.000	0.038	9.26	0.3704	1040.42	1040.70	0.283
	2.000	0.074	7.90	0.6318	1079.80	1079.54	-0.261
	4.001	0.138	7.53	1.2051	1150.90	1149.81	-1.088
	5.996	0.193	7.98	1.9139	1212.49	1210.41	-2.079
	7.988	0.242	8.09	2.5847	1265.96	1263.69	-2.275

T /°C	Molality / mol/kg	Mass Fraction	Water Volume / mL	Solute Mass / gr	Density from literature / kg/m³	Density exp / kg/m³	Density Residual / kg/m³
	NaSCN						
20	0.500	0.039	8.00	0.3242	1017.84	1018.25	0.413
	1.000	0.075	8.00	0.6485	1036.47	1037.16	0.689
	2.001	0.140	8.00	1.2978	1071.03	1071.79	0.764
	4.000	0.245	8.00	2.5946	1130.65	1130.28	-0.366
	6.001	0.327	8.00	3.8920	1180.21	1180.34	0.126

2nd measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Volume / mL	Solute Mass / gr	Density from literature / kg/m ³	Density exp / kg/m ³	Density Residual / kg/m ³
	NaOH						
20	0.000	0.000	0.00	0.00	998.20	998.20	0.000
	0.200	0.008	23.91	0.1913	1006.88	1006.86	-0.019
	0.300	0.012	15.04	0.1805	1011.17	1010.89	-0.283
	0.500	0.020	16.33	0.3265	1019.66	1019.46	-0.204
	0.700	0.027	6.90	0.1933	1028.07	1027.78	-0.289
	0.999	0.038	12.02	0.4805	1040.39	1040.25	-0.141
	2.000	0.074	10.92	0.8734	1079.81	1079.12	-0.688
	4.000	0.138	5.18	0.8282	1150.87	1147.18	-3.692
	6.002	0.194	10.51	2.5232	1212.66	1207.11	-5.551
	7.949	0.241	5.23	1.6628	1264.99	1252.83	-12.158

T /°C	Molality / mol/kg	Mass Fraction	Water Volume / mL	Solute Mass / gr	Density from literature / kg/m ³	Density exp / kg/m ³	Density Residual / kg/m ³
	NaSCN						
20	0.500	0.039	6.01	0.2438	1017.86	1017.92	0.064
	1.001	0.075	6.00	0.4869	1036.51	1035.55	-0.960
	1.994	0.139	6.02	0.9733	1070.80	1069.98	-0.824
	3.992	0.245	6.01	1.9450	1130.41	1128.74	-1.672
	5.993	0.327	8.01	3.8919	1180.04	1179.13	-0.910

3rd measurements

T /°C	Molality / mol/kg	Mass Fraction	Water Volume / mL	Solute Mass / gr	Density from literature / kg/m³	Density ^{exp} / kg/m³	Density Residual / kg/m³
	NaOH						
20	0.000	0.000	0.00	0.00	998.20	998.20	0.000
	0.201	0.008	22.39	0.1799	1006.92	1007.25	0.334
	0.300	0.012	11.57	0.1391	1011.19	1011.60	0.409
	0.500	0.020	16.33	0.3265	1019.66	1020.17	0.506
	0.700	0.027	12.31	0.3449	1028.06	1028.69	0.628
	1.001	0.038	8.37	0.3351	1040.45	1041.36	0.907
	2.000	0.074	8.28	0.6622	1079.83	1080.29	0.463
	3.998	0.138	8.51	1.3608	1150.79	1150.78	-0.007
	6.000	0.194	8.03	1.9271	1212.60	1213.91	1.312
	7.991	0.242	8.39	2.6816	1266.04	1265.28	-0.763

T /°C	Molality / mol/kg	Mass Fraction	Water Volume / mL	Solute Mass / gr	Density from literature / kg/m³	Density exp / kg/m³	Density Residual / kg/m³
	NaSCN						
20	0.501	0.039	8.03	0.3260	1017.87	1018.64	0.769
	0.999	0.075	8.02	0.6496	1036.44	1037.22	0.778
	1.998	0.139	8.01	1.2977	1070.94	1072.09	1.151
	3.988	0.244	8.03	2.5959	1130.29	1131.40	1.105
	6.001	0.327	8.00	3.8922	1180.22	1179.66	-0.561

1st measurements

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution density from literature / mPa*s	Solution exp density / mPa*s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN			
Mixed Solution									
20	0.500	0.000	0.0196	0.0000	0.1567	0.0000	7.8433	1019.6497	1019.46
	0.500	0.100	0.0194	0.0079	0.1567	0.0639	7.8433	1025.4700	1023.96
	0.500	0.201	0.0193	0.0157	0.1567	0.1275	7.8433	1029.3096	1027.76
	0.500	0.302	0.0191	0.0234	0.1567	0.1919	7.8433	1033.1596	1031.99
	0.500	0.500	0.0188	0.0382	0.1567	0.3179	7.8433	1040.5833	1039.44
	0.500	0.700	0.0186	0.0527	0.1567	0.4450	7.8433	1047.9288	1046.41
	0.500	1.000	0.0181	0.0736	0.1567	0.6356	7.8433	1058.6827	1057.11
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	1040.4123	1040.70
	1.000	0.100	0.0382	0.0078	0.3077	0.0626	7.6923	1046.2644	1044.59
	1.000	0.200	0.0379	0.0154	0.3077	0.1249	7.6923	1049.8616	1048.25
	1.000	0.301	0.0376	0.0230	0.3077	0.1880	7.6923	1053.4713	1051.95
	1.000	0.500	0.0370	0.0375	0.3077	0.3118	7.6923	1060.4565	1058.39
	1.000	0.700	0.0365	0.0518	0.3077	0.4367	7.6923	1067.3758	1066.21
	1.000	1.000	0.0357	0.0723	0.3077	0.6236	7.6923	1077.4957	1076.20
20	2.001	0.000	0.0741	0.0000	0.5927	0.0000	7.4073	1079.8450	1079.54
	2.001	0.102	0.0735	0.0076	0.5927	0.0610	7.4073	1085.8085	1083.36
	2.001	0.201	0.0730	0.0149	0.5927	0.1206	7.4073	1088.9418	1086.74
	2.001	0.301	0.0725	0.0221	0.5927	0.1805	7.4073	1092.0657	1091.21
	2.001	0.501	0.0714	0.0362	0.5927	0.3008	7.4073	1098.2639	1096.48
	2.001	0.700	0.0704	0.0499	0.5927	0.4204	7.4073	1104.3270	1102.90
	2.001	1.000	0.0689	0.0698	0.5927	0.6005	7.4073	1113.2738	1111.74
20	3.001	0.000	0.1072	0.0000	2.4538	0.0000	20.4429	1116.6159	1116.15

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution density from literature / mPa*s	Solution exp density / mPa*s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN			
	3.000	0.101	0.1064	0.0072	0.8571	0.0583	7.1429	1122.5853	1119.35
	3.000	0.200	0.1056	0.0143	0.8571	0.1160	7.1429	1125.3312	1122.11
	3.001	0.301	0.1049	0.0213	0.8573	0.1743	7.1427	1128.1221	1125.28
	3.000	0.500	0.1034	0.0349	0.8571	0.2897	7.1429	1133.4858	1131.63
	3.000	0.700	0.1020	0.0483	0.8571	0.4056	7.1429	1138.8337	1136.85
	3.000	1.001	0.0999	0.0676	0.8571	0.5796	7.1429	1146.7229	1145.59
20	5.997	0.000	0.1935	0.0000	1.5478	0.0000	6.4522	1212.5230	1210.41
	5.997	0.100	0.1922	0.0065	1.5478	0.0524	6.4522	1218.6884	1214.30
	5.997	0.200	0.1910	0.0129	1.5478	0.1046	6.4522	1220.4591	1216.21
	5.997	0.300	0.1898	0.0192	1.5478	0.1569	6.4522	1222.2298	1219.75
	5.997	0.500	0.1873	0.0317	1.5478	0.2615	6.4522	1225.7597	1223.16
	5.997	0.700	0.1850	0.0438	1.5478	0.3661	6.4522	1229.2725	1227.60
	5.997	1.001	0.1816	0.0614	1.5478	0.5235	6.4522	1234.5212	1232.86

2nd measurements

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution density from literature / mPa*s	Solution exp density / mPa*s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN			
Mixed Solution									
20	0.500	0.000	0.0196	0.0000	0.1569	0.0000	7.8431	1019.6724	1019.88
	0.500	0.101	0.0195	0.0079	0.1569	0.0641	7.8431	1025.5049	1024.15
	0.500	0.201	0.0193	0.0157	0.1569	0.1277	7.8431	1029.3443	1028.04
	0.500	0.300	0.0192	0.0233	0.1569	0.1909	7.8431	1033.1225	1031.88
	0.500	0.500	0.0189	0.0382	0.1569	0.3179	7.8431	1040.6054	1039.28

T/°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution density from literature / mPa*s	Solution exp density / mPa*s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN			
	0.500	0.700	0.0186	0.0527	0.1569	0.4453	7.8431	1047.9677	1046.56
	0.500	1.001	0.0182	0.0737	0.1569	0.6367	7.8431	1058.7651	1057.25
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	1040.4123	1040.42
	1.001	0.100	0.0382	0.0078	0.3079	0.0625	7.6921	1046.2868	1044.95
	1.001	0.200	0.0379	0.0154	0.3079	0.1249	7.6921	1049.8896	1048.40
	1.001	0.301	0.0376	0.0229	0.3079	0.1875	7.6921	1053.4706	1052.08
	1.001	0.501	0.0370	0.0376	0.3079	0.3124	7.6921	1060.5174	1059.27
	1.001	0.701	0.0365	0.0518	0.3079	0.4371	7.6921	1067.4247	1066.34
	1.001	1.000	0.0357	0.0723	0.3079	0.6238	7.6921	1077.5327	1076.47
20	2.000	0.000	0.0741	0.0000	0.5925	0.0000	7.4075	1079.8093	1079.12
	2.000	0.100	0.0735	0.0075	0.5925	0.0602	7.4075	1085.7305	1083.59
	2.000	0.200	0.0730	0.0148	0.5925	0.1204	7.4075	1088.8958	1087.11
	2.000	0.300	0.0724	0.0220	0.5925	0.1803	7.4075	1092.0201	1090.48
	2.000	0.501	0.0714	0.0362	0.5925	0.3006	7.4075	1098.2190	1096.78
	2.000	0.701	0.0704	0.0500	0.5925	0.4208	7.4075	1104.3130	1102.93
	2.000	1.000	0.0689	0.0699	0.5925	0.6008	7.4075	1113.2551	1113.27
20	3.000	0.000	0.1071	0.0000	0.8571	0.0000	7.1429	1116.5851	1115.86
	3.000	0.100	0.1064	0.0072	0.8571	0.0580	7.1429	1122.5759	1119.43
	3.000	0.200	0.1056	0.0142	0.8571	0.1156	7.1429	1125.3171	1122.04
	3.000	0.302	0.1049	0.0214	0.8571	0.1746	7.1429	1128.1058	1124.94
	3.000	0.500	0.1034	0.0350	0.8571	0.2898	7.1429	1133.4951	1130.73
	3.000	0.701	0.1020	0.0483	0.8571	0.4059	7.1429	1138.8521	1135.97
	3.000	1.000	0.0999	0.0675	0.8571	0.5791	7.1429	1146.7050	1144.55
20	6.000	0.000	0.1936	0.0000	1.5483	0.0000	6.4510	1212.6132	1210.42

T / °C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution density from literature / mPa*s	Solution exp density / mPa*s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN			
	6.000	0.100	0.1923	0.0065	1.5483	0.0523	6.4510	1218.7757	1213.02
	6.000	0.200	0.1911	0.0129	1.5483	0.1045	6.4510	1220.5458	1215.50
	6.000	0.300	0.1898	0.0192	1.5483	0.1569	6.4510	1222.3194	1218.40
	6.000	0.501	0.1874	0.0317	1.5483	0.2620	6.4510	1225.8652	1221.41
	6.000	0.700	0.1851	0.0438	1.5483	0.3660	6.4510	1229.3568	1225.47
	6.000	1.000	0.1817	0.0614	1.5483	0.5231	6.4510	1234.5942	1232.05

3rd measurements

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution density from literature / mPa*s	Solution exp density / mPa*s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN			
Mixed Solution									
20	0.500	0.000	0.0196	0.0000	0.1570	0.0000	7.8430	1019.6847	1020.61
	0.500	0.101	0.0195	0.0079	0.1570	0.0640	7.8430	1025.5112	1024.35
	0.500	0.200	0.0193	0.0157	0.1570	0.1272	7.8430	1029.3265	1028.00
	0.500	0.300	0.0192	0.0233	0.1570	0.1909	7.8430	1033.1347	1031.80
	0.500	0.500	0.0189	0.0382	0.1570	0.3180	7.8430	1040.6232	1039.14
	0.500	0.700	0.0186	0.0527	0.1570	0.4453	7.8430	1047.9795	1046.47
	0.500	1.000	0.0182	0.0737	0.1570	0.6361	7.8430	1058.7433	1057.12
20	1.000	0.000	0.0385	0.0000	0.3077	0.0000	7.6923	1040.4123	1042.68
	1.001	0.100	0.0382	0.0077	0.3079	0.0623	7.6921	1046.2752	1044.70
	1.001	0.200	0.0379	0.0154	0.3079	0.1248	7.6921	1049.8838	1048.55
	1.001	0.300	0.0376	0.0229	0.3079	0.1871	7.6921	1053.4478	1052.22
	1.001	0.501	0.0370	0.0376	0.3079	0.3123	7.6921	1060.5118	1059.20

T /°C	Molality / mol/kg		Mass Fraction		Solute Mass / g		Water Mass / g	Solution density from literature / mPa*s	Solution exp density / mPa*s
	NaOH	NaSCN	NaOH	NaSCN	NaOH	NaSCN			
	1.001	0.700	0.0365	0.0518	0.3079	0.4368	7.6921	1067.4082	1066.35
	1.001	1.000	0.0357	0.0723	0.3079	0.6238	7.6921	1077.5327	1076.52
20	2.000	0.000	0.0741	0.0000	0.5925	0.0000	7.4075	1079.8093	1080.51
	2.000	0.100	0.0735	0.0075	0.5925	0.0603	7.4075	1085.7358	1083.79
	2.000	0.200	0.0730	0.0148	0.5925	0.1201	7.4075	1088.8801	1087.21
	2.000	0.301	0.0724	0.0221	0.5925	0.1808	7.4075	1092.0460	1090.77
	2.000	0.501	0.0714	0.0362	0.5925	0.3006	7.4075	1098.2190	1097.01
	2.000	0.701	0.0704	0.0500	0.5925	0.4208	7.4075	1104.3130	1103.46
	2.000	1.000	0.0689	0.0698	0.5925	0.6005	7.4075	1113.2404	1112.27
20	3.000	0.000	0.1071	0.0000	0.8571	0.0000	7.1429	1116.5851	1116.38
	3.000	0.101	0.1064	0.0072	0.8571	0.0583	7.1429	1122.5902	1119.27
	3.000	0.201	0.1056	0.0143	0.8571	0.1164	7.1429	1125.3550	1122.83
	3.000	0.300	0.1049	0.0212	0.8571	0.1735	7.1429	1128.0540	1125.32
	3.000	0.500	0.1034	0.0349	0.8571	0.2897	7.1429	1133.4905	1130.57
	3.000	0.701	0.1020	0.0483	0.8571	0.4058	7.1429	1138.8475	1136.37
	3.000	1.001	0.0999	0.0676	0.8571	0.5798	7.1429	1146.7364	1144.81
20	6.000	0.000	0.1936	0.0000	1.5483	0.0000	6.4510	1212.6132	1210.84
	6.000	0.101	0.1923	0.0066	1.5483	0.0529	6.4510	1218.7961	1213.52
	6.000	0.201	0.1910	0.0130	1.5483	0.1053	6.4510	1220.5730	1215.66
	6.000	0.300	0.1898	0.0192	1.5483	0.1570	6.4510	1222.3228	1217.90
	6.000	0.500	0.1874	0.0317	1.5483	0.2615	6.4510	1225.8484	1221.39
	6.000	0.700	0.1851	0.0438	1.5483	0.3663	6.4510	1229.3668	1225.19
	6.000	1.001	0.1817	0.0614	1.5483	0.5234	6.4510	1234.6042	1232.73

