

Experimental and Theoretical Considerations of Electrolyte Conductivity in Glucose Alkaline Fuel Cell

Lea Mor¹, Zeev Rubin²

¹Department of Biotechnology Engineering, Ort Braude College, Karmiel, Israel

²Physics Unit, Ort Braude College, Karmiel, Israel

Email: leamor@braude.ac.il, rubin_zeev@yahoo.com

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ABSTRACT

The paper focuses on the conductivity of the fuel cell electrolyte in a membraneless glucose-fueled alkaline fuel cell. The electrolyte conductivity is interpreted using simple physical models, considering either the empirical behavior of the solution's viscosity, or the consideration of ions and molecules colliding in solutions. The conductivity is expressed as a function of KOH and glucose concentrations. The physical properties of the species (*i.e.* radii, thermal velocity) and the chemical equilibrium constant of the reaction that glucose undergoes in an alkaline solution can be estimate by comparing the experimental results with the theory.

Keywords: Alkaline; Fuel Cell; Glucose; Electrolyte; Conductivity; Collision

1. Introduction

Fuel cell resistivity is one of the factors determining cell efficiency. The resistivity affects fuel cell power [1-3], efficiency and stability [4]. The resistivity of a glucose-fuelled alkaline fuel cell was previously found to depend linearly on the glucose concentration [5] dissolved in a constant KOH concentration of 0.35 M, and was shown to be seven times higher than the resistivity of a KOH-only solution [6].

The increase in resistivity of the alkaline glucose, or other carbohydrate's solutions, was also considered previously, to be due to the complexation of alkali ions with reducing and nonreducing carbohydrates [7].

The present study aimed to explain the relationship between the glucose and KOH concentrations and the changes in the fuel cell electrolyte resistivity over time.

The overall fuel cell resistance is defined as the sum of the resistances of the bulk solution (electrolyte), the diffusion layer [8] and the electrodes [3]. The electrolyte resistance was assumed to be larger than the electrodes' resistance [3]. Various authors have tried to develop theoretical and experimental methods for estimating the various components of fuel cell resistance [3,8]. Among these methods were: a micro-scale model for predicting contact resistance [8], a monodimensional simulation model of the overall electrical resistance due to ohmic losses and polarization phenomena in stationary conditions, assuming equilibrium of the chemical reactions [9], a dynamic model, based on experimental characteriza-

tions of the anode and cathode equivalent resistance [3], and a transient model of the cathode catalyst layer [10].

This paper focuses on using experimental and theoretical considerations to study electrolyte conductivity of an alkaline glucose solution.

Two theories, explaining the behaviour of conductivity as function of glucose and KOH concentration, will be presented at the third section. The experimental behaviour of conductivity in a test tube and in alkaline fuel cell will be introduced in the fourth section. Analysis of the experimental results using both theories is discussed in the 5th section.

2. Materials and Methods

The solutions investigated here were of glucose-KOH with various concentrations ranging from 0.05 M to 1 M for both the glucose and KOH. The solutions were prepared shortly before use by dissolving the glucose in water or in KOH solutions. KOH concentration was monitored before and during the experiments by titration with potassium biphthalate (Carlo Erba). It should be noted that measurement by titration with biphthalate gives the total OH⁻ concentration, but nonetheless, this measurement is referred to as the change in KOH concentration. The KOH is the main source of the ions in solution, and is responsible for the solution conductivity. The experiments include test tubes and open/closed circuit's fuel cell.

Test tube—A test tube of 50 ml volume, contained 25

ml glucose-KOH solution. The conductivity was measured in the test tube.

Open/closed circuit's fuel cell—The membraneless alkaline fuel cell, (purchased from Hong Kong University [11,12]), constructed with one reservoir including both the fuel glucose and the electrolyte KOH solution. Its cathode came into contact with the ambient air and its anode, with incorporated platinum particles, was immersed in fuel/electrolyte (25 ml glucose-KOH) solution. For close circuit's fuel cell experiments, the circuit was closed with a load resistance of 10 Ω . For electrolyte conductivity measurements the solution were temporarily transferred to a 50 ml test tube; after getting the conductivity result, the solution was returned to the fuel cell.

The temperature in both experiments was maintained in a water bath adjusted to 21 C.

Conductivity and KOH concentrations were measured every 20 minutes for the first five hours, and once a day for up to five days. Conductivity was measured with a Cyberscan 510 conductivity meter. Samples for HPLC analysis over time were conducted using an isocratic pump using eluting solvent acetonitrile:water 80:20 (v/v).

3. Theory

Equation (1) expresses the electrolyte conductivity, derived from the ions present in the electrolyte solution before and during the fuel cell operation, Using simplified kinetic theory considerations [13,14].

$$\sigma = e \sum_i |z_i| u_i c_i \quad (1)$$

here σ is the conductivity and e is the elementary charge. The summation index i refers to the ions that take part in electricity conduction in solution: K^+ and OH^- generated from the electrolyte, H^+ generated from the water, and g^- produced during a chemical reaction of glucose in the alkaline solution. z_i is the signed ionic charge of the i^{th} ion, c_i is its concentration and u_i is its mobility coefficient, defined by:

$$u_i = v_{di} / E. \quad (2)$$

v_{di} is the drift velocity of the ions, induced by the electric field E . Equation (1) is composed of two parts: properties of the solution (e.g. concentrations and viscosity) and specific parameters of the ions (e.g. charge, velocity and effective size). The concentration of the ions derived from the electrolyte and water were assumed, for this presentation: The concentrations of K^+ , OH^- and KOH are identical, and the concentration of H^+ is negligible compared to the concentration of OH^- . The concentration of the ions produced during the chemical reactions (i.e. endiolate ($C_6H_{11}O_6^-$) or gluconate ($C_6H_{11}O_7^-$)) were assumed to be $a_{g^-} = k_{g^-} c_g c_{OH^-}$, where K_{g^-} was the equi-

librium constant for the ions generated from the glucose in the alkaline solution.

3.1. Conductivity-Viscosity Empirical Evaluation

The mobility coefficient is often expressed by the Stokes-Einstein equation [13]:

$$u_i = \frac{|z_i| e}{6\pi\eta r_i}. \quad (3)$$

Here, η is the solvent viscosity (in this case, of an aqueous glucose solution) and r_i is customarily called the (hydrated) ionic radius of the i^{th} ion. Equation (3) appears to describe a sphere with charge $z_i e$, moving at a terminal velocity v_{di} under the influence of an electrical field E and a Stokes-Law drag force (i.e., a drag force in a creeping flow around a sphere). However, this is not the physical picture: Given that the ions and the water (and glucose) molecules are of similar orders of magnitude, the ions are not surrounded by an inter-molecular fluid and no drag whatsoever acts on them. Consequently, Equation (3) is used as an empirical approximation, rather than as a statement of a force balance on an ion. In this context, $1/\eta$ represents the notion that sluggishly flowing solvents do hinder the mobility of the ions in them and $e/(6\pi r_i)$ is a coefficient, characteristic of the i^{th} ion. Consequently, substituting Equation (3) into Equation (1), and introducing all the definitions and assumptions, yields:

$$\frac{1}{\sigma} = \frac{6\pi\eta}{e^2 \left(\frac{c_{OH^-}}{r_{OH^-}} + \frac{c_{K^+}}{r_{K^+}} + \frac{K_{g^-} c_g c_{OH^-}}{r_{g^-}} \right)} \quad (4)$$

We now introduce the concept of normalized conductivity:

$$\bar{\sigma} = \sigma / [KOH] \quad (5)$$

Since KOH is the major contributor to the conductivity, this normalization reveals the changes in conductivity undergone by the other factors in the solution, with the exception of KOH.

Equation (4) leads to:

$$\frac{1}{\bar{\sigma}} = \frac{6\pi\eta}{e^2 N_A \left(\frac{1}{r_{OH^-}} + \frac{1}{r_{K^+}} + \frac{K_{g^-} c_g}{r_{g^-}} \right)}. \quad (6)$$

This completes the inclusion of the Stokes-Einstein Equation (3) in expression (1). Empirical expressions (4) and (6) can be used for processing the measured conductivity values. These two expressions include the assumption in expression (1) of constant u_i values, which may prove suitable when only the ionic concentrations affect

the conductivity, and the assumption in expression (3), which can handle some of the glucose concentration's effect, because η varies with the glucose concentration in the solvent [15].

3.2. Conductivity-Collision Evaluation

Let us now define the acceleration under the influence of an electric field, v_{di}/τ_i . Introducing Newton's second law gives:

$$E = v_{di} m_i / q_i \tau_i. \quad (7)$$

τ_i is the mean average characteristic time of successive collisions between the i^{th} ion and glucose molecules $\tau_i = 1/c_g \sigma_{gi} v_i$. σ_{gi} is the cross-section of collision $\pi(r_i^2 + r_g^2)$ [16], v_i is the thermal velocity of the i^{th} ion in the solution, c_g is the glucose concentration and r_i and r_g are the hydrated radii of the ions and glucose in the solution. Introducing the definitions and assumptions of Equation (1) yields:

$$\frac{1}{\sigma} = \frac{c_g}{e^2 \left(\frac{c_{KOH}}{\sigma_{gK^+} v_{K^+} m_{K^+}} + \frac{c_{KOH}}{\sigma_{gOH^-} v_{OH^-} m_{OH^-}} + \frac{K_g c_g c_{KOH}}{\sigma_{gg^-} v_{g^-} m_{g^-}} \right)} \quad (8)$$

Introducing the normalized conductivity definition (5) to Equation (8) leads to

$$\frac{1}{\bar{\sigma}} = \frac{c_g}{e^2 \left(\frac{1}{\sigma_{gK^+} v_{K^+} m_{K^+}} + \frac{1}{\sigma_{gOH^-} v_{OH^-} m_{OH^-}} + \frac{K_g c_g}{\sigma_{gg^-} v_{g^-} m_{g^-}} \right)} \quad (9)$$

The theoretical expressions (8) and (9) can be used for processing of measured conductivity values. They include the assumption in equation (1) of constant u_i values, which may prove suitable when the colliding species are hard, not interacting spheres, as in dilute gases [17].

4. Results

4.1. Conductivity Measurements in the Test Tube

Since electrolytes play a major part in fuel cell resistance, the first step in studying glucose-fuelled alkaline fuel cell resistance changes over time is to investigate the instantaneous conductivity of an alkaline glucose solution in a test tube. The conductivity of KOH solutions (0.05 M to 1 M) with various glucose concentrations, up to 1 M, was measured shortly after preparation. The resistivity

(the reciprocal of conductivity) for each initial KOH concentration was plotted against the glucose concentration in **Figure 1**. It can be seen that for each initial KOH concentration the glucose, a nonionic compound, increased the solution's resistivity (decreased the solution conductivity). To discern the changes in the solutions' conductivity caused by factors other than KOH—which is the major contributor to the conductivity—the normalized resistivity ρ (reciprocal of normalized conductivity) was plotted against the glucose concentration in **Figure 2**. Once all the graphs of the different KOH concentrations were combined into one graph, the data indeed showed that the glucose concentration was responsible for the conductivity changes. To demonstrate the effect of time on fuel cell electrolyte conductivity, the conductivity was monitored in three solutions, representing high (0.8 M), medium (0.25 M) and low (0.05 M) KOH concentrations. The data shows that normalized resistivity was linear with the glucose concentration for both instantaneous and subsequent measurements (after 3 - 4 days).

4.2. Changes in Normalized Conductivity over Time

Since the changes in normalized conductivity over time is function of both KOH concentration and solution conductivity, both KOH concentration and solution conductivity were measured over time in two solutions, representing high (0.35 M KOH, 1 M glucose) and low (0.1 M KOH, 0.1 M glucose) concentrations. Two experiments per solution were conducted.

4.2.1. KOH Concentration Changes over Time

The mean KOH concentration decreased with time both in the test tube and in the open/closed fuel cell during the five days of the experiment (**Figure 3**).

The concentration changes in the test tube were higher

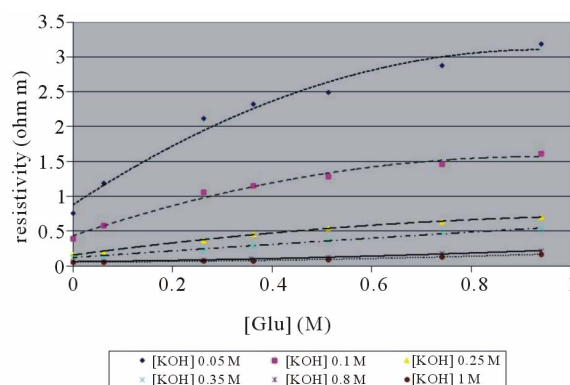


Figure 1. The resistivity of glucose solution in KOH, for each KOH concentration, is plotted against glucose concentration. Measurements were taken in the test tube shortly after preparing the solution.

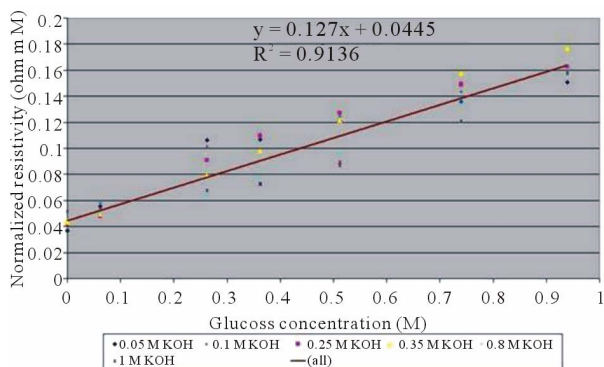


Figure 2. Normalized resistivity, as a function of glucose concentration at various KOH concentrations. Measurements were taken in the test tube shortly after preparing the solution.

when the KOH concentration was high (0.35 M) as opposed to low (0.1 M). Significant changes between the KOH concentration in the test tube and in the fuel cell, for both KOH concentrations, were evident. The decrease in KOH concentration over time in the low (0.1 M) KOH concentration resulted from the reaction of KOH with CO_2 , which already existed, and was continuously dissolving in the water. In the high (0.35 M) KOH concentration, the higher decrease may result from the isomerization reactions of glucose, catalyzed by the high KOH concentration, producing endiolate ions. In the fuel cell, the fuel cell electrode can, in addition, catalyzes electrochemical reaction, in both KOH concentrations, yielding gluconate ions, which contributed to the lower KOH concentration in the fuel cell [18].

4.2.2. Conductivity Changes over Time

4.2.2.1. Test Tube

The reduction of the solution's mean conductivity measured in the test tube depended on the amount of time that had elapsed (Figure 4). For the first few hours, the effect was high, and thereafter remained stable for the next few days. Comparing changes in the KOH concentration (Figure 3) and conductivity (Figure 4), led to the following inferences: The rate of KOH concentration changed during the first day; at a concentration of 0.1 M KOH, it was similar to the rate of the conductivity change. In opposition, when the KOH concentration was 0.35 M, the rate of the KOH concentration change was much larger than the rate of the conductivity change. This effect can be clearly expressed by the normalized resistivity ($[\text{KOH}]/\sigma$) (Figure 5). The normalized resistivity in solution was almost constant at 0.1 M KOH, whereas it dropped significantly in a concentration of 0.35 M KOH, because of the reaction took place in a solution, catalyzed by a high KOH concentration. Ionic compounds generated by this reaction contributed to the conductivity, and kept it almost constant in spite of the

reduction in OH^- concentration.

4.2.2.2. Fuel Cell

The situation was different in the fuel cell. Here, under both KOH concentrations, despite the high reduction in KOH concentration over time (Figure 3), the reduction in conductivity is less profound (Figure 4). Consequently, the normalized resistivity (Figure 5) decreased over time for both KOH concentrations. This can be explained by generation of ionized compounds g^- by the electrochemical reaction.

4.2.3. Normalized Resistivity Changes with Glucose Concentration

The normalized resistivity in two glucose concentrations are shown in Figure 6(a) for the test tube, and in Figure

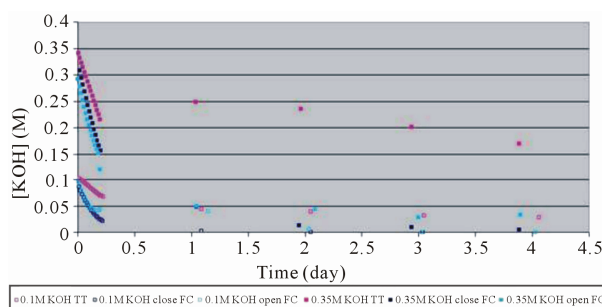


Figure 3. KOH concentration changes in open/closed fuel cell, compared to test tube.

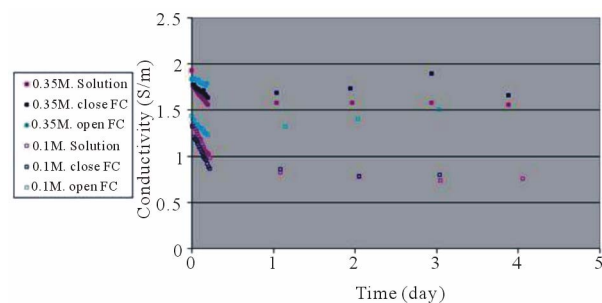


Figure 4. Conductivity changes in open/closed fuel cell, compared to test tube.

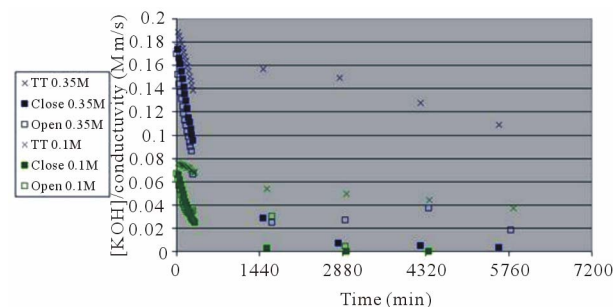


Figure 5. Normalized resistivity ($[\text{KOH}]/\text{conductivity}$) changes in open/closed fuel cell, compared to test tube.

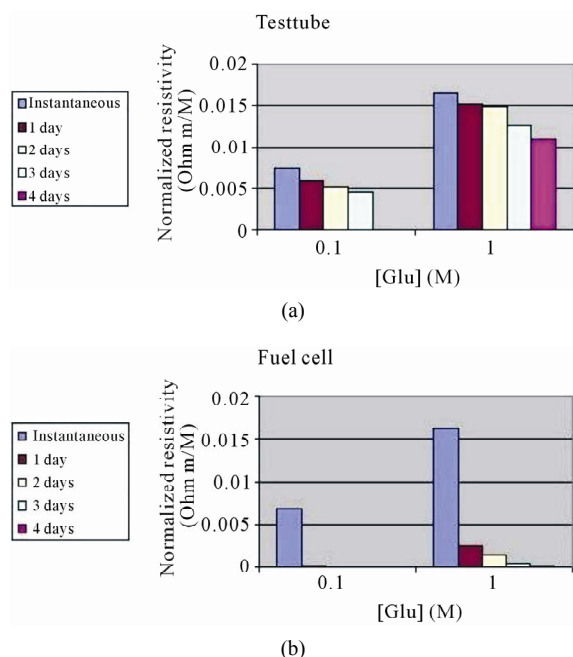


Figure 6. Effect of time on normalized resistivity under two glucose concentrations, [Glu]. (a) Test tube; (b) Fuel cell.

6(b) for the fuel cell. It can clearly be seen that in the test tube, there was a slow and graduated decrease in normalized resistivity in both glucose concentrations (high and low). In the fuel cell, there was a dramatic drop in normalized resistivity beginning from the first day onward.

4.3. HPLC Analysis of the Glucose-KOH Solution

To observe the chemical changes in an alkaline glucose solution, HPLC analysis of glucose (0.1 M) in KOH (0.1 M) was done (Figure 7). Two dominant species (Rt 9.6–9.8 and 13), had no significant change in concentration over time, were evident in the test tube (Figure 7(b)). In the fuel cell, these species concentrations change (either increase or decrease over time). In the fuel cell, there was an additional species (Rt 10.5), which increased in concentration over time (Figure 7(a)). Other species (Rt 7.1, 8 and 8.9), having low concentration, exist in both test tube and fuel cell with no significant changes over time. These findings prove that there are differences between the chemical reactions occurring in the fuel cell (the electrochemical reaction and isomerization) and in test tube (only isomerization).

5. Discussion

5.1. Analysis of the Experimental Results Using the Conductivity-Viscosity Empirical Evaluation for Low Glucose Concentration

Assuming low glucose concentrations

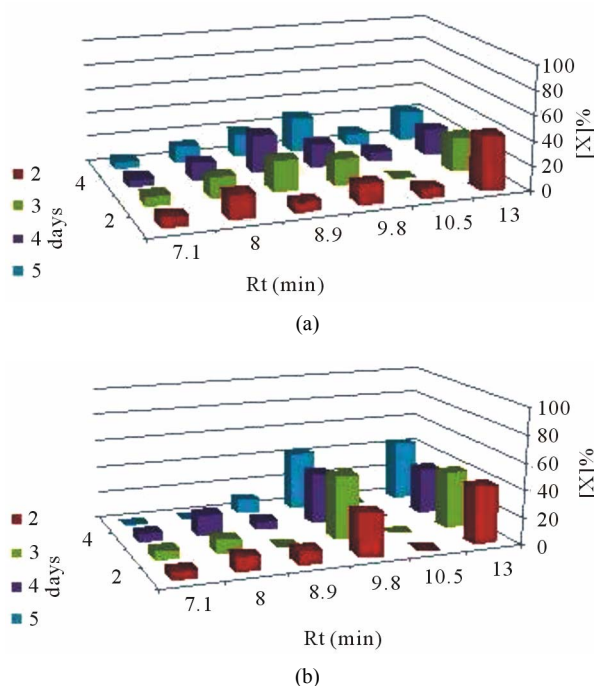


Figure 7. Changes in the alkaline glucose solution in: (a) fuel cell, (b) test tube. [Glu] 0.1 M, [KOH] 0.1 M, elution solvent AcN:H₂O 80:20.

$$\left(\frac{1}{r_{\text{OH}^-}} + \frac{1}{r_{\text{K}^+}}\right) \cdot \left(r_{\text{g}^-} / K_{\text{g}^-}\right) \gg c_{\text{g}}, \quad (10)$$

and assuming

$$r_{\text{OH}^-} = r_{\text{K}^+} = r_i, \quad (11)$$

the normalized conductivity (Equation (6)) is simplified to:

$$\frac{1}{\bar{\sigma}} = \bar{\rho} = \frac{3\pi\eta r_i}{e^2 N_A}. \quad (12)$$

The viscosity has a linear dependence on the glucose concentration (Equation (13)) [15]:

$$\eta = 0.7308E-3[G] + 0.9307E-3, \text{ Pas}. \quad (13)$$

Exploring (Equation (13)) and (Equation (12)), it is evident that the normalized resistivity is a linear function of the glucose concentration, in agreement with the experimental results (Equation (14)), Figure 2):

$$\bar{\rho} = 0.1271[G] + 0.0445. \quad (14)$$

By fitting the experimental results (Equation (14)) to Equation (12,13), two values of r_i were found: 2.84E–10 m (2.84 Å), and 0.78 Å. The error in estimating r_i here is approximately 57% relative to its mean value. Both values of r_i are in agreement with the range of (1.37–10) Å obtained from the estimated hydrated and unhydrated ion radii [19]. The minimal value of r_i , taking into account the ion radii in crystals and solutions [15], cannot be below 1.37 Å. Hence, the model predicted that

the ions were in hydrated form.

The experimental results showed a drop in normalized resistivity over time in a fuel cell (**Figure 5**). The model explains the experimental time effect by assuming increase in g^- over time. This finding agrees with the HPLC data showing the production of a chemical species in the fuel cell, which was not present in the test tube.

5.2. Analysis of the Experimental Results Using the Conductivity-Collision Equation Assuming a Low Glucose Concentration

For low glucose concentrations,

$c_g \ll \sigma_{g^-} \nu_{g^-} m_{g^-} / K_{g^-} \sigma_{gi} \nu_i m_i$, Equation (9) can be simplified and expressed in Equation (15), in terms of the normalized resistivity as a function of glucose concentration.

$$\bar{\rho} = \frac{c_g}{e^2 \left(\frac{1}{\sigma_{gK^+} \nu_{K^+} m_{K^+}} + \frac{1}{\sigma_{gOH^-} \nu_{OH^-} m_{OH^-}} \right)}. \quad (15)$$

Equation (15) shows that the normalized resistivity is linear with the glucose concentration, thus agreeing with the experimental results (**Figure 2**). However, it does not describe the resistivity of the pure KOH solution, in the absence of glucose, which is expressed by the intercept in **Figure 2**.

5.3. The Viscosity

The viscosity expression (Equation 16) was evaluated using the mobility definition (Equation (2)) in both the models (Equations (3) and (7)).

$$\eta = \sum \frac{m_i \sigma_{ji} \nu_i}{6\pi r_i} \cdot c_g \quad (16)$$

Equation (16) shows that the solution's viscosity, an empirical characteristic of the solution, is basically a function of parameters influencing the collision in the solution, and is linear with the solute (glucose) concentration. This explains the mechanism by which the viscosity influences the solution resistivity.

6. Conclusion

Glucose increases the resistivity of a KOH solution. Two simple physical models were used to explain the contribution of glucose to fuel cell resistivity, based on the solution viscosity or collisions between ions and glucose molecules in solution. The simple collision model proposed here is also applicable to solutions. It predicts the species parameters. Glucose degrades under alkaline conditions in fuel cells, which can increase fuel cell resistivity.

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Nomenclature

Latin Symbols

C_i : Molar concentration of the i^{th} chemical species, kmol/m³

c_i : concentration of the i^{th} chemical species, 1/m³

e : Elementary charge, 1.602×10^{-19} C

K_{g-} : Equilibrium constant for ions generated from glucose, m³/kmol

m_i : Ion mass, kg

r_i : Length coefficient (also called the “hydrated ion radius”) for the i^{th} ion, m

v_{di} : Drift velocity, m/sec

v_{mi} : v_i is a thermal velocity, m/sec

u_i : Mobility coefficient of the i^{th} ion, m²·sec⁻¹·V⁻¹

z_i : Signed ionic charge, expressed as the number of elementary charges

Greek symbols

η : Viscosity, Kg·m⁻¹·sec⁻¹

ρ : Electrical resistivity, $\Omega \cdot \text{m}$

$\bar{\rho}_{\text{KOH}} = 1/\bar{\sigma}_{\text{KOH}}$: Normalized resistivity, $\Omega \cdot \text{m} \cdot \text{kmol}/\text{m}^3$

σ : Electrical conductivity, $\Omega^{-1} \cdot \text{m}^{-1}$

σ_{ij} : Cross section collision between two species i with j , m²

$\bar{\sigma}_{\text{KOH}} = \frac{\sigma}{[\text{KOH}]}$: Normalized conductivity, $\Omega^{-1} \cdot \text{m}^{-1} \cdot \text{m}^3/\text{kmol}$

τ_{ij} : Time between collisions, sec

Subscripts

g, G : Glucose

I : The i^{th} collide species

J : The j^{th} species collided with

Abbreviations

Å: 10⁻¹⁰ m

g^- : Glucose ions (C₆H₁₁O₆)⁻

mol: Gram molecular weight

Rt: Retention time, min

TT: Test tube

[X]: Molar concentration (M) of the X chemical species, kmol/m³