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Performance evaluation of the Anion exchange membrane based Water electrolysis*

Ronit Kumar Panda^{1a}, Guillaume Serre^{1a}, Frédéric Fouda Onana^{1a}, Yann Bultel^{1b} and Pascal Schott^{1a}

Abstract— Anion exchange membrane (AEM) electrolysis is the process for hydrogen generation using electricity. In this article, a 1D steady state model was introduced and elaborated to determine the performance of the AEM Electrolyser. In particular, the role of addition of Potassium Hydroxide (KOH) and its overall impact on the Multiphysics phenomenon inside the membrane electrode assembly was investigated. The simulation and experiments showed a significant reduction of the ion exchange / transport, ionic resistance as well as an improvement of the reaction kinetics at the electrodes upon addition of KOH to the pure water feedstock. The modelling efforts also resulted in a better understanding of the several Multiphysics phenomena that account for the kinetics and mass-transfer losses in the electrolyser. In particular, we also assess the fluid, geometric and flow parameters for the AEM electrolyser and the extent of their impact on the device performance.

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I. INTRODUCTION

Hydrogen has been accepted as the efficient and effective energy carrier from all sustainable resources. In the quest to obtain efficient renewable and clean vectors of energy, we shift our focus towards Anion Exchange Membrane Water Electrolysers (AEMWE). These electrolysers unlike their predecessors operate in the alkaline environment without using noble catalysts hence reducing the cost of electrolytic process. The main difference between alkaline and AEM electrolysis is the replacement of the conventional diaphragm with an AEM in alkaline water electrolysis. In the frame of the NewEly European project, several studies deal with the development and testing of components for the next generation AEMWE electrolyser, such as the membrane, electrodes and stack design. The goal is to mainstream the economic adoption of green hydrogen and to bring down the CAPEX cost by using non-noble catalysts. In the recent times, studies have been conducted to better understand the electrode-electrolyte interface as well as the effect of various electrode materials on the hydrogen production and its corresponding efficiency.[1] The mathematical model allowed us to better explore the benefits as well as detrimental effect of addition of KOH over a wide range of concentrations. Following our analysis, we decided to continue our operation at 0.01M- 0.1M to ensure a higher performance (reduce the electrolysis voltage) and sufficient durability for an extended operation of the electrolyser. [2] [3]

II. AEM ELECTROLYSER MODEL

A. Introduction

A schematic for the AEMWE is illustrated in the **Figure 1**.

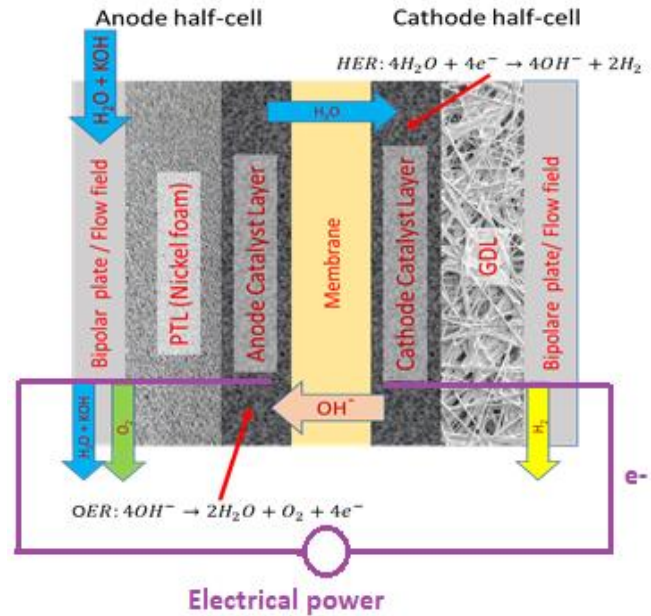


Figure 1 Schematic of the Anion exchange membrane based Water electrolysis

The water electrolyte is supplied to the anode compartment and the water molecules are transported through the membrane, while the cathode side remains dry. The water molecules pass through the polymer membrane and undergo reduction at the cathode terminal to generate hydrogen. The power supplied from the external circuit is used to create an electrical potential difference at the interface of the electrode layers and the electrodes. In above figure, we can see one circuit with two types of current: the electron one induced by the generator and the ionic one that closes the whole circuit and that is induced by the electrochemical reactions in the electrodes. The electrons can move in the bipolar plate, the PTL, the GDL and the electrodes but not in the membrane nor in the ionomer. The hydroxyl ions OH^- move only in the ionomer in the electrodes and in the membrane. In the electrodes, at the catalyst surface, the Oxygen evolution reaction (OER) takes place at the anode while at the cathode the Hydrogen evolution reaction (HER) produces the hydrogen gas and OH^- using the electrons. As a result, these two reactions, the electrical circuit is closed in the catalyst layers and hydrogen is produced. In order to minimise the electrical power supplied to the device, several improvement tracks are explored such as maximising the utilisation of the available catalytic sites at the electrodes.

The objective of this paper is to:

1. Formulate the model that describe the performance of an AEM electrolyser (electrochemical, fluid and ion transport), describe the Multiphysics code that is the receptacle of these models and its validation on experimental results.
2. Using this code, study the influence of the KOH concentration on the electrolyser performance.

The principal parts of the AEM electrolyser as seen in Figure 1 are:

- The Membrane Electrode Assembly (MEA) comprises the solid electrolyte (AEM), ionomer, anode and cathode catalyst layers. Both the Oxygen and Hydrogen evolution reactions at the anode and cathode respectively require electrocatalysts to overcome the kinetics of the reaction. Ionomers are binders that assist in creating transport pathways between the membrane and the reaction sites in the catalyst layer.
- Gas Diffusion Layers at cathode and sinter (Nickel foam) at anode. These Porous Transport Layers have to conduct electricity (electrons) as well as gas, water and heat. On the anode side, since water has to be achieved to the electrode, the surface of the sinter is hydrophilic (contact angle lower than 90°). On the cathode side, the GDL is hydrophobic to remove the water (contact angle $>90^\circ$).
- Flow-field plates (indicated as bipolar plate in **Figure 1**) which partition each cell in the electrolyser stack and which are machined with a liquid channel for circulation of the water.
- Flowing liquid electrolyte (Water + KOH) produces hydrogen at the cathode. The KOH is added to water in order to improve its ionic conductivity and increase the efficiency of the electrolytic reactions.

B. Implementation of the Model in MATLAB

The model structure is based on the Bond Graph theory with Resistance elements for transport equations and Capacity elements for molar and energy balance equations. The schematic for AEMWE modelling is illustrated in **Figure 1**. A material balance is carried out to calculate the mole fractions of the species undergoing electrochemical reactions at the electrodes. In concurrence with the material balance, we also determine the resultant molar flux of water through the membrane (AEM). The electrochemical model assumes concentration-dependent Tafel kinetics at each electrode and the ionic conductivity of the membrane and ionomer as a function of the KOH concentration at the anode. The reaction current density variation inside the electrode due to the addition of KOH at the inlet is also considered in our model. [4–6] The local conditions at the electrodes are considered to calculate the electrochemical response of the cell. It is being implemented in the MePHYSTO code developed by CEA in MATLAB/ SIMULINK [7]. The code is discretized by a meshing 1D through plane and 1D in-plane providing the capability to calculate the molar flux of liquid and gases along the channels and through the various components.

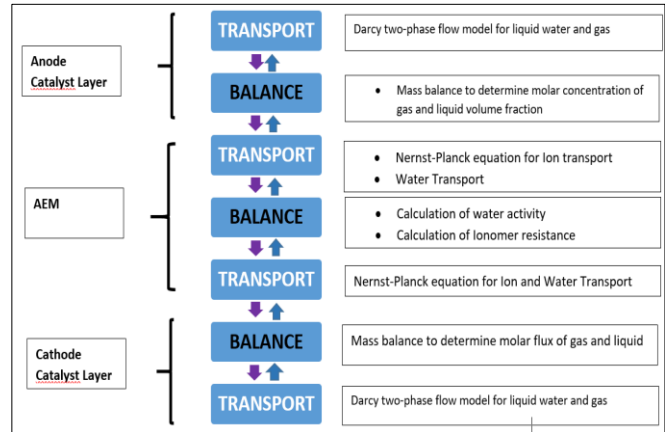


Figure 2 Structure of the electrolyser model in this paper

C. Assumptions

- The in-plane transport is modelled in the channel components, neither in the PTL nor in the membrane as it is assumed that the convection in the channels dominates the diffusion along the membrane and the Porous Transport Layer (PTL). The convection of liquid and gases through plane are modelled.
- Each catalyst layer is treated like an interface. This questionable and provisional hypothesis come from the origin of MePHYSTO: the code was developed for PEM fuel cell where this thickness is negligible compared to the GDL one.
- The chemical potential of the species is calculated at the electrodes assuming a constant temperature and thermodynamic equilibrium through the thickness of the electrode. But, these thermodynamic parameters can vary between meshes along the plane.
- At steady state and in equilibrium, the electrochemical potential of hydroxide ions in the membrane and the solution are equal.
- Local effect of hydroxide concentration at the electrodes are accounted within the activity coefficient of the hydroxide ions according to Debye-Huckel theory.
- Dilute solution theory is considered for the Nernst-Planck equations.
- All gases (including vapour) are assumed to follow ideal gas law.
- No fuel or oxidant crossover are considered.
- The temperature of the electrodes are calculated by the thermal balance (enthalpy equations) in the overall cell accounting for the heat reaction.

Figure 3 highlights the model skeleton described in this paper. **Table 1** highlights the input parameters in the model and several electrochemical parameters that are used in our model. All the parameters in this paper unless mentioned are in SI units.

III. MATHEMATICAL MODELLING

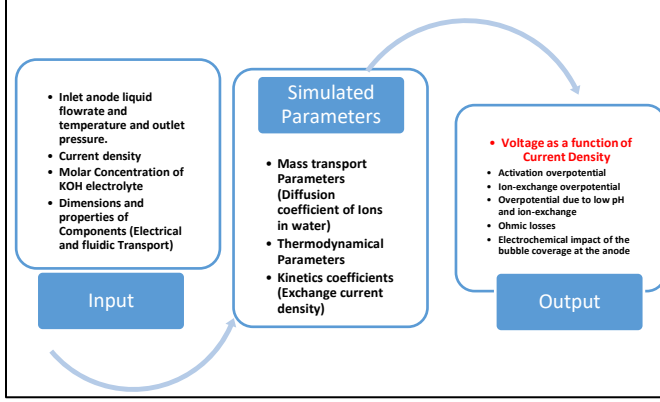


Figure 3 Performance model for AEM Water electrolysis

A. Abbreviations and Acronyms

Table 1 Model Parameters

Symbol	Name	Value
α_a, α_c	Anodic and cathode charge transfer coefficient (HER)	0.5
α_a, α_c	Anodic and cathode charge transfer coefficient (OER)	1
K	Thermodynamic Constant	$-0.009 \text{ J (mol.K)}^{-1}$
n	Constant in the bubble coverage model	0.33
$\kappa_i, 0.1M$	Membrane ionic conductivity with 0.1M KOH[8]	75 mS/cm
$\kappa_i, diwater$	Membrane ionic conductivity with pure water	14 mS/cm

C. Performance Model

In an electrolytic cell, the existence of overpotential implies the cell requires more energy than thermodynamically required for a non-spontaneous reaction. This additional voltage is required to overcome both the reaction kinetics at the electrodes and Ohmic resistance of the electrolyte and components of the electrolyser. The cell voltage (U_{cell}) is calculated using the following equation:-

$$U_{cell} = U_0 + K(T - T_{ref}) + \frac{RT}{nF} \ln \frac{a_{H_2} a_{O_2}^{0.5}}{a_{H_2O}} + \eta_{cat} + \eta_{an} + \eta_{ohm} + \eta_{ion-exchange} + \eta_{pH} + \eta_{membrane} + \eta_{bubble} \quad (1)$$

where η_{cat} , η_{an} are the activation overpotential at the cathode and anode respectively. η_{pH} is the overpotential due to the pH gradient through the membrane. $U_0 = -\frac{\Delta G^0}{nF}$ is the standard reversible potential. The overpotential due to the Ohmic

resistance of the electrolyser components (GDL and CL), η_{ohm} and $\eta_{ion-exchange}$ refers to the overpotential resulting from the ion transport between the ionomer and liquid electrolyte. $\eta_{membrane}$ refers to the overpotential due to the resistance in transport of ions in the AEM. [9]

The gases and water activities are given by:

$$a_{H_2} = \frac{P_{H_2}}{P_{ref}}, a_{O_2} = \frac{P_{O_2}}{P_{ref}}, a_{H_2O} = \frac{P_{H_2O}}{P_{saturation}} \quad (2)$$

P_{ref} is the reference pressure (1 bar), $P_{saturation}$, the water saturation pressure and P_i , the partial pressure of the gases. The activity of water is determined as the ratio between water vapour partial pressure and saturated vapour pressure ($P_{saturation}$) since the water activity of saturated vapour is the same as that of water in the liquid phase.[10]

1.1. Ion and liquid electrolyte transport model through the membrane

The model includes laws for the calculation of the overpotential due to the transport of ions through the AEM. The involved phenomena are electro-migration, diffusion of ion and streaming potential driven by the chemical potential gradient of water. It is expressed through the Nernst-Planck equation:

$$N_{OH^-,M} = \frac{-k_{OH^-} - \gamma_{OH^-}}{z_{OH^-} F} \nabla \eta_{membrane} - \frac{-\xi_{OH^-} k_{OH^-} - \gamma_{OH^-}}{(z_{OH^-} F)^2} \nabla \mu_{H_2O} - D_{OH^-,M} \nabla C_{OH^-,M} \quad (3)$$

Where k_{OH^-} , z_{OH^-} , ξ_{OH^-} , γ_{OH^-} and μ_{H_2O} are respectively the conductivity, charge, electroosmotic coefficient, mole fraction of hydroxyl ions within the membrane-ionomer and the chemical potential of water. The ion transport with the pressure gradient results in a net charge transport i.e. streaming current denoted by the chemical potential term. Conversely, fluid flow that occurs due to potential difference is referred to as electro-osmotic flow. $D_{OH^-,M} \nabla C_{OH^-,M}$ refers to the diffusion component due to the ion transport through the membrane. The water flow rate through the membrane is expressed as a sum of flow due to diffusion and electro-osmosis. The model has been elaborately described in **Equation 4**. The electro-osmotic drag is directly related to the flux of hydroxide ions migrating to the anode from the cathode which can be expressed as:

$$N_{H_2O,M} = \frac{D_{Water}^M}{\delta_{an}} \left[\left(\frac{\rho_{Water} T_{cathode}}{M_{water}} + \frac{\delta_{cat} n_{cat}^{water}}{D_{eff}^{cat}} \right) - \left(\frac{\rho_{Water} T_{anode}}{M_{water}} - \frac{\delta_{an} n_{an}^{water}}{D_{eff}^{an}} \right) \right] + \frac{1 \cdot eod}{F} \quad (4)$$

It is also to be mentioned that the values of electro-osmotic drag have been fitted to the experimental values and is related to the water uptake and temperature of the membrane. In the electrolyte, the flux of OH^- results from diffusion and migration:

$$N_{OH^-,E} = z_{OH^-} F m_{OH^-} C_{OH^-,E} \nabla \eta_{electrolyte} - D_{OH^-,E} \nabla C_{OH^-,E} \quad (5)$$

$$\Delta \eta_{ion-exchange} = \frac{1}{i_{cell}} \delta (R_{exchange} F (\eta_{membrane} - \eta_{electrolyte})) \quad (6)$$

Where m_{OH^-} , $D_{OH^-,E}$, $C_{OH^-,E}$ are respectively the ionic mobility, the diffusion coefficient, the ionic concentration of hydroxyl ions within the liquid electrolyte. δ refers to the thickness of the electrodes. $R_{exchange}$ is the ion exchange mass flux inside the electrolyte flowing through the electrolytic cell. This flux is nil for DI water, thus, ionic flux exists through the catalyst/ionomer. The addition of KOH allows supplementary ion transport pathways through the liquid electrolyte. As a result, overpotential due to this ion-exchange within the solution, is created at the membrane-liquid electrolyte interface.[11]

For diffusion of charged particles, the electrical mobility equation is obtained in accordance with the Einstein equation as follows:

$$D = \frac{\mu_q k_B T}{q} \quad (7)$$

Where D is the Diffusion coefficient, μ_q is the electrical mobility of charged particles, q is the electric charge of particle.[12]

The ionic current density is a function of the flux density of the anion:

$$i_{OH,liq^-} = z_{OH^-} F N_{OH,liq^-} \quad (8)$$

$R_{exchange} =$

$$k_{exchange} \left(\frac{C_{OH^-}^{membrane} \exp\left(\frac{z_i F (\eta_{membrane} - \eta_{electrolyte})}{RT}\right)}{C_{OH^-}^{electrolyte}} \right) - \quad (9)$$

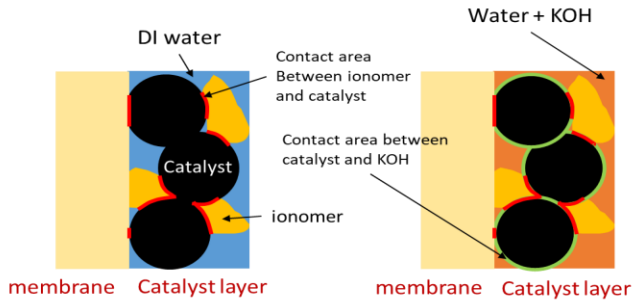


Figure 4 Effect of KOH Concentration on the ionic conductivity of the AEMWE Components

1.2 Electrochemical Active Surface Area (ECSA)

For the context of our work, ECSA is defined here as the ratio between the available active sites for reaction at the catalyst surface to the projected area of the catalyst. It is a dimensionless quantity.[13] In the domain of the fuel cells and electrolysis, numerous approaches have been undertaken to calculate or measure the ECSA. Liu et al. [13] undertook the approach of multiple interfaces to explain the enhancement due to addition of KOH. In our work, we assumed a transition-state theory complex and a single rate limiting reaction at each electrode, which resulted in equations 10, 11 and 12.

$$\eta_{cat,an} = \frac{RT}{\alpha_c F} \operatorname{arcsinh}\left(\frac{i}{2 i_{0,cat/an}}\right) \quad (10)$$

As mentioned above $i_{0,an}$ and $i_{0,cat}$ in electrochemical reactions are analogous to the rate constant of a chemical reaction.

$$i_{0,cat} = i_0 \frac{a_{an} ECSA}{a_c} \exp\left(\frac{-E_c}{RT} \left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right) \quad (11)$$

$$ECSA = \frac{a_c L_c}{\text{Geometric area of electrodes}} \quad (12)$$

i_0 is the reference exchange current density per unit catalyst area (at reference temperature and pressure, typically 25°C and 101.325 kPa), a_{cat} the anode/cathode activity, E_c is the activation energy (J), a_c = catalyst specific area, L_c = catalyst loading, P_r = reactant partial pressure, kPa. P_r^{ref} = Reference pressure, (100kPa), T = Operating Temperature and T_{ref} = Reference temperature (298.15K).[14]

1.3 Gas bubbles at the electrodes

In the electrolyser, the bubbles, attached to the surface of electrodes, insulate a portion of the electrodes and make them inefficient. We accommodate this effect by reducing the portion of electrode surface area using the material and liquid properties. These released bubbles cause an additional overpotential at the anode.

$$\eta_{bubble} = \frac{RT}{\alpha_a F} \ln\left(\frac{1}{1-\theta}\right) \quad (13)$$

$$\theta = B \left(\frac{J}{A/m^2}\right)^n \quad (14)$$

It is assumed that the bubble coverage of the cathode is negligible due to faster kinetics of HER and hence we neglect the contribution on the cathode side.[15]

Ohm's law determines the ionic current density:

$$i_{electronic} = -\sigma_{electronic, \frac{electrode}{liquidelectrolyte}} \nabla \eta_{ohm} \quad (15)$$

Where $\sigma_{electronic}$ refers to the effective conductivity of the electrode layers. Gas bubbles and their growth and release can further increase the Ohmic resistance. In the literature, there are several equations to describe this effect of electrolyte solution occupied by bubbles. In our work, we use the equation derived by Maxwell to calculate the electrical conductivity of the electrolyte solution in AEMWE and taking into account the gas volume fraction (p) given as [16]:

$$\frac{\sigma_{electronic,bubble}}{\sigma_{electronic,electrode/liquidelectrolyte}} = \frac{1-p}{1+0.5p} \quad (16)$$

1.4 Influence of alkaline media

This overpotential is due to the intrinsic reaction kinetics in the electrodes and arise due to the lower concentration of hydroxide ions.[17] This effect is largely reduced when a higher concentration of KOH is added to the pure water feedstock at the inlet. The mobile hydroxide ions generated at the cathode and consumed at the anode and the AEM is completely charge compensated by the ion K+. [18][19]

$$\eta_{pH} = \frac{RT}{F} \ln(10)(pH_{cathode} - pH_{anode}) \quad (17)$$

2 RESULTS AND DISCUSSION

The model has been validated using experimental data obtained at CEA. They are based on a 2-cm² single cell test bench where the membrane was PSEBS and the MEA was made with CCS electrodes that consists in NiFe and Moca

catalysts from CENmat on the anode and cathode respectively. PTL are made of Nickel foam as anode diffusion layer and a PEM fuel cell GDL (H23C7 from Freudenberg) as cathode GDL.[20]

In this work, we have provisionally used a simplified version of the code, using 0D approach. Thus, the operating conditions such as the pressure, temperature and concentrations are imposed directly on the electrochemical models.

We have:

1. Validated the polarisation curve and calculated electrical response of the electrolyser with the experiments conducted within the framework of the NEWELY European project. The inlet concentration of KOH were pure water feed and 0.1M KOH. All the materials were developed to achieve the objective of $1\text{A}/\text{cm}^2$ at 2V in low concentrated KOH solution (0.1 M KOH). The exchange current densities requested by the model were found by fitting Tafel kinetics slope to the polarisation curves
2. Performed sensitivity analysis for DI water and 0.1 M KOH at $0.1\text{ A}/\text{cm}^2$. (**Figure 6 and Figure 7**)

The **Figure 5** illustrates the sensitivity to the KOH concentration of the various contributions that participate to the potential. It also facilitates the reaction at the cathode while enhancing the ion exchange at the anode. It is to be noted that the ion-exchange overpotential is set to nil for DI water in our model. Upon increasing the amount of KOH concentration, the pH overpotential at the anode significantly reduces. We also observe a drastic reduction in the membrane ionic resistance as the KOH raises the ionic conductivity by a factor of two. The OER kinetics remains a bottleneck at lower hydroxide concentrations.

The polarisation curves in **Figure 6** and **Figure 7** show that the model bears a very good agreement with that of the experiments. The membrane overpotential with the addition of KOH is halved mostly arising from the reduction in the Ohmic resistance of the electrodes and a significant improvement in the intrinsic anode reaction kinetics.

The higher values of exchange current density signify the faster reaction kinetics resulting in lowering the activation losses. The overpotential at the anode is four times that at the cathode. This is a result of the sluggish kinetics of the oxygen evolution reaction at the anode and the relatively faster kinetics at the cathode. The cathode kinetics loss is small in all cases due to the non-sluggish and faster kinetics of HER with the use of CENmat MoCa as catalyst.

OPERATIONAL PARAMETERS	
Parameter	Value
Membrane thickness (microns)	180
ACL thickness (microns)	10
CCL thickness (microns)	5
Operating Temperature in Kelvin	323
Operating Pressure in bar	1

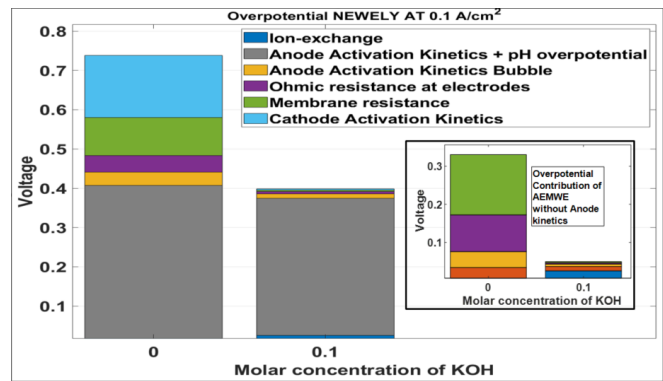


Figure 5 Applied voltage breakdown in AEMWE as a function of hydroxide concentration

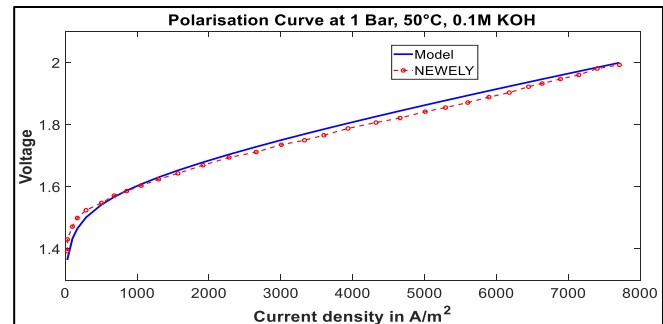


Figure 6 Polarisation curve for NEWELY materials based MEA for 0.1M KOH at 50°C

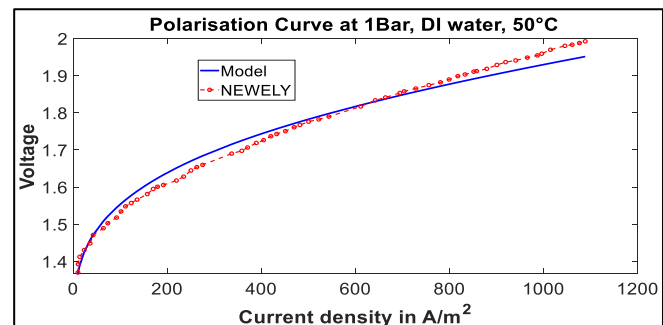


Figure 7 Polarisation curve for NEWELY materials based MEA for DI water feedstock at 50°C

3 CONCLUSION

A mathematical model of an anion exchange membrane-based electrolyser cell was developed to analyse the performance. This model includes the physical, electrochemical laws associated with the various components of the electrolyser: the anode, the cathode, and the membrane. Compared to previous published models for performance calculation, our model considers the impact on ECSA of the bubble effect, which makes the model more realistic. In the future, as a continuation of the performance model, a 1D simulation will be performed. Using the performance model, further tests and modelling efforts are planned in the future to further investigate the role of bubbles and their long-term impacts on the durability of the AEMWE systems. Optimising the Multiphysics phenomenon at the Membrane Electrode assembly as well as the device components would ensure a sustained higher performance of the electrochemical device.

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