

Multi-impedance Distribution of Relaxation Times Applied to Predicting Fuel Cell Stack Operating State: A Theoretical and Experimental Study

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The distribution of relaxation times (DRT) converts the impedance data into a simple peak signal, which can be used to quickly identify the operating state of the fuel cell. However, the DRT peak obtained from a single impedance value cannot accurately define the internal reaction process in the fuel cell. In this paper, the variation pattern of the operating state of the fuel cell stack is determined by analysing the peaks of each single cell. The results show that, for DRT, a regularization parameter (λ) of 10⁻⁵ can produce the optimal peak, whereas the change in the shape of its peak can be better used to analyse the operating state of the stack. Moreover, the operating state of the stack can be adjudged and predicted by analysing the changes of P1, P2 and P3 peaks of DRT. Additionally, the reasons for lower performance were also analysed.

Keywords: PEMFC; Multi-impedance; Distribution of Relaxation Times; Regularization Parameter

1. INTRODUCTION

Proton exchange membrane fuel cells (PEMFCs) have the advantages of green zero emissions, high power density and high efficiency and have been widely used in new energy vehicles, backup power, distributed power generation and other fields [1-4]. However, due to the problems associated with bipolar plates, gas diffusion layer (GDL), gaskets and other materials and assembly methods in fuel cell stack [5-8], the performance of each cell is different because of uneven pressure distribution. Detecting the reaction process of each unit in the stack is of great significance for predicting fuel cell stack operating state.

The monolithic consistency of the stack is mainly related to the water/gas/electricity transmission state in the electrode of each unit during operation, so it is necessary to evaluate the stack by online monitoring technology [9-12]. The online monitoring technology of the stack mainly includes the polarization curve, current interruption, AC impedance and cyclic voltammetry [13, 14]. Among them, electrochemical impedance spectroscopy (EIS) can determine the operating status of water and gas in the stack without disturbing the normal operation of the stack [15, 16]. EIS uses a small sinusoidal voltage (or current) signal with variable frequency that perturbs the test system to stimulate an impedance response, thus obtaining the corresponding system parameter information. The data obtained by the EIS test include the real and imaginary parts of the impedance, phase angle, modulus and other related information. The above data need to be analysed mathematically to evaluate the physical and chemical changes inside the stack. At present, the most commonly used method for EIS data processing is generally the model method, including equivalent circuit models (ECM) and physical-based impedance models [17]. ECM uses a series of common circuit elements, such as resistance, inductance and capacitance, to simulate various polarization processes that may exist in fuel cell operation. Physical-based impedance models can predict and interpret EIS data under different working conditions by coupling a series of control equations. However, the ECM-based EIS data analysis method has limitations. Different circuit models can often represent the same impedance expression, so the circuit model needs to pre-set several polarization processes of the fuel cell according to prior experience. In Nyquist plots, due to the close time scale of partial polarization processes, multiple arcs in the impedance spectrum overlap into one. These factors limit the identification and recognition of various reaction processes inside the fuel cell to a certain extent, making the physical meaning of ECM unclear.

The distribution of relaxation time (DRT) is a new data processing method that has been proposed in recent years. DRT represents the quantification of the relaxation process in the impedance frequency response process. It combines countless R||C circuits to represent the impedance spectrum and converts the EIS data into simple peak signals through mathematical methods such as the Fourier transform method, regularization method and genetic algorithm. In 2017, Heinzmann applied DRT for the first time to detect PEMFC single cells and analysed the contribution of various polarization processes during their operation [18]. Reshetenko studied the mass transfer impedance of a low-Pt PEMFC cathode using DRT [19]. Moreover, Bevilacqua studied the DRT characteristics under changing anode operating parameters (such as nitrogen dilution and CO poisoning) [20]. Schiefer investigated the low-frequency inductance characteristics under different operating conditions using DRT [21]. The DRT calculations contain some important input parameters, including frequency range, the number of points per decade, impedance values, and regularization parameter. The selection of these parameters has important influence on the results of DRT calculations. Among them, the most valuable parameter is the regularization parameter (λ), which smoothens and avoids artificial DRT peaks [22]. However, all the above studies are based on the analysis results of a single impedance value for either a single cell or the entire stack, while DRT conversion involves the selection of regularization parameters, which will have a greater impact on the DRT peak. Due to the difference in the selection of parameters in the above studies, the correlation between the reaction process inside the fuel cell and the peak signal cannot be accurately defined.

In order to solve the abovementioned problems, more comparative tests are needed to obtain the

optimal parameters. One feasible method is to obtain the optimal peak signal by synchronously testing each single cell in the stack and then comparing and analysing the test results of each single cell. However, there is no relevant report to date on the analysis of the EIS data of each single cell of the stack using DRT technology.

In this paper, the distribution of the relaxation time method is used to convert the EIS data of each single cell in the stack under different operating conditions, such as clamping pressure, stoichiometry, relative humidity (RH) and back pressure into a simple peak signal. The optimal regularization parameter is obtained by analysing the peak shape of the DRT of each single cell. The variation pattern of the operating state and the peak is then obtained by combining the polarization curve data of each cell. The schematic of the process is shown in Figure 1.

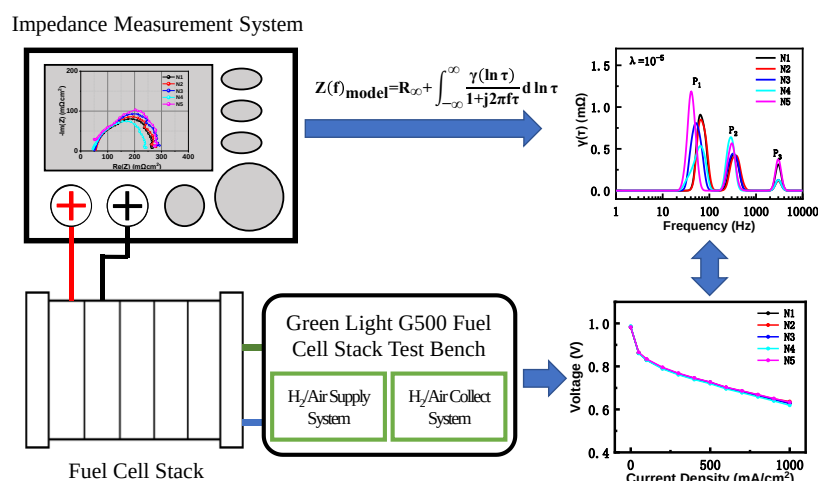


Figure 1. Schematic diagram of the stack test and data conversion process

2. EXPERIMENTAL

2.1 Assembly of the stack

The stack used in the experiment was a graphite plate stack developed by Wuhan University of Technology (WUT), China. The stack was composed of five single cells. The membrane electrode assemblies (MEAs) used in the stack were a commercial standard load type produced by WUT New Energy Co., Ltd., China. The stack controlled the GDL compressibility in the stack through different clamping pressures, measured the stress distribution of the GDL under different compression ratios through tactile and pressure measurement system, drew the stress-strain curve, and then measured at different clamping pressures (3 t, 4 t, 5 t, and 6 t). The stress distribution of each unit of the stack was used to estimate the compression rate. Fig. S1a (Supporting Information; SI) is the stress-strain curve of the GDL, whereas Fig. S1b shows the pressure distribution of the stack under the clamping pressures of 3 t, 4 t, 5 t, and 6 t. The average compression rates of the GDL were 17.7%, 21.55%, 25.2% and 26.8% for the clamping pressures of 3 t, 4 t, 5 t and 6 t, respectively.

2.2 Performance test

The test equipment mainly consisted of the GreenLight G500 test bench, the KIKUSUI AC impedance test system (voltage monitoring: KFM2151, impedance meter: KFM2150, electronic load: PLZ1004 W, amplifier: PLZ2004 WB) and a Tekscan tactile and pressure measurement system.

During the test process, the G500 was responsible for the adjustment of supply and collecting data of gas, cooling water, temperature, and back pressure. Moreover, the AC impedance system was used for monitoring the voltage/current, adjusting the input AC signal of the stack and collecting the feedback signals. It ensures that the AC signal will not destroy the stability, linearity and causality of the stack or cause significant changes. In addition, it ensures an appropriate signal-to-noise ratio to avoid small amplitudes that may get affected by noise, which will affect the interpretation of the results. Experiments have found that there is no significant difference in the impedance spectroscopy between 5% and 10% of the AC amplitude. Therefore, 5% AC amplitude was used in the current work. The test frequency used in this work was 0.7Hz-3000Hz. In order to improve the reliability of the collected data, a moving average using 16 periods was chosen.

Before the impedance test, the stack was stabilized at the test point for 10 min to ensure that the stack reached a stable state. In this work, two points of 500 mA/cm² and 1000 mA/cm² were selected for the AC impedance test. In order to ensure that the stack can avoid local gas starvation and obtain enough fuel at a low current density, the theoretical gas flow required at 112 A was selected as the initial minimum flow rate.

2.3. DRT calculations

The DRT method can separate time constants in the fuel cell reaction process so that each polarization process can be displayed more clearly [22]. The impedance data processing method abstracted the electrochemical system into ohmic resistance and countless R||C series forms, as shown in Figure 3(a). Each R||C electrochemical system was used to represent the relaxation process at different time scales, and its specific expressions are given by Eq. (1).

$$Z(f)_{model} = R_{\infty} + Z_{pol}(f) = R_{\infty} + \int_0^{\infty} \frac{g(\tau)}{1 + j2\pi f\tau} d\tau \quad (1)$$

where R_{∞} is the ohmic resistance, $Z_{pol}(f)$ is the polarization impedance, $\tau = RC$ is the time constant of a single R||C element, and $g(\tau)$ is the distribution function of the relaxation time. Since the frequency of the impedance spectrum is generally divided into logarithms, it is necessary to convert the variables of Equation (1) into logarithms, so Eq. (1) can be rewritten as Equation (2).

$$Z(f)_{model} = R_{\infty} + Z_{pol}(f) = R_{\infty} + \int_{-\infty}^{\infty} \frac{\gamma(\ln \tau)}{1 + j2\pi f\tau} d \ln \tau \quad (2)$$

where $\gamma(\ln \tau) = \tau g(\tau)$. The purpose of the DRT method is to obtain an estimate of $\gamma(\ln \tau)$ by deconvoluting $\gamma(\ln \tau)$ from Eq. (2).

In this work, the regularization method is used for the approximation of $\gamma(\ln \tau)$. Regularization is a common method for the calculation and solution of this type of ill-posed problem [23].

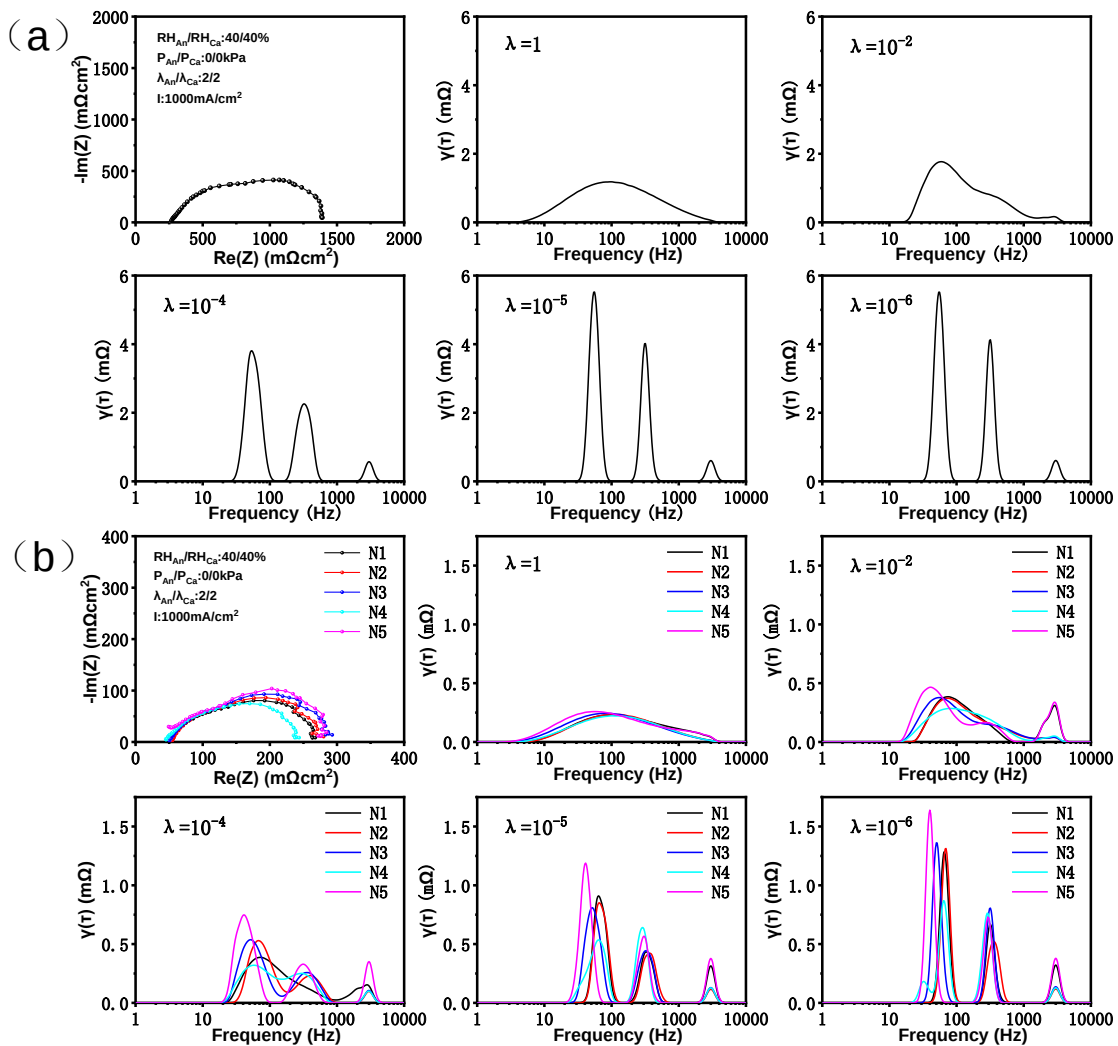


Figure 2. (a) Typical circuit model of the DRT method; (b) EIS test data of the stack and the characteristic peaks of DRT when λ is 10^{-4} and 10^{-5} , respectively; (c) EIS test data of each single cell in the stack and the characteristic peaks of DRT when λ is 10^{-4} and 10^{-5} , respectively.

The expression for this regularization process is given by Equation (3).

$$\sum_{n=1}^N [y_n - h(x_1, x_2, \dots, x_N, f_n)]^2 + \lambda P(x_1, \dots, x_N) \quad (3)$$

where y_n is the experimental data corresponding to the sampling point, f_n is the frequency corresponding to the sampling point, $h(x_1, x_2, \dots, x_N, f_n)$ corresponds to the fitting function of the model, and λ and $P(x_1, \dots, x_N)$ are the regularization parameter and the penalty term, respectively (the purpose of which is to optimize the stability of the calculated results), x_N is the weight of the fitting function, and Eq. (4) is the expression for solving the DRT through regularization.

$$S(x) = \|Z_{\text{exp}}(f_n) - \mathbf{A}\mathbf{X}\|_2^2 + \lambda P(\mathbf{X}) \quad (4)$$

In Eq. (4), $Z_{\text{exp}}(f_n)$ is the experimental value sampled at the f_n frequency point, $\mathbf{A}$ is the matrix obtained by the $Z(f)$ model, and $\mathbf{X}$ is the N -dimensional weight vector $[x_1, x_2, \dots, x_N]$.

The appropriate λ value is determined by comparing the effects of different λ values on the DRT characteristic peaks of the entire stack and each single cell. The test results are shown in Figures 2(a) and 2(b). For the impedance data of the entire stack, different λ values can produce characteristic peaks,

whereas the choice of λ is uncertain. For the impedance data of each single cell in the stack, due to the inconsistent operating states of each single cell, the peaks were prone to overlapping when λ lied within the range of 10^{-4} . Furthermore, the characteristics of the peaks at various frequencies were not clear, so the choice of this value was not conducive to analysing the test results. When λ lied within the range of 10^{-5} , the peaks of each single cell in the stack were clear and changed regularly. Therefore, this value was a more suitable choice for analysis. Due to this reason, all DRT data processing in this paper adopted a λ value of 10^{-5} .

3. RESULTS AND DISCUSSION

The fuel cell stack used in this work was a 1.5 kW stack with five single cells (active area was 300 cm^2) and GDL compression was 21.55%. More details about the setup can be found in the Supporting Information. Usually, the frequency area of the fuel cell impedance test can be divided into two parts: a low-frequency area below 10^3 Hz , which represents the mass transfer process and the charge transfer process; and a high-frequency area above 10^3 Hz , which represents the ohmic resistance [21]. Some previous studies have reported similar results on the attribution and allocation of DRT characteristic peaks [23,24]. Based upon the changes in DRT, three typical characteristic peaks can be obtained in the low-frequency (marked as P1 and P2) and high-frequency regions (marked as P3). P1 is related to the water/gas mass transfer state, which can be used to assess whether the fuel cell is flooded or is experiencing lack of gas. P2 is related to the charge transfer state, which can be used to assess the effective active area or O_2/H^+ concentration of the catalyst's surface. Moreover, P3 peak is related to the electronic and ionic resistances of the fuel cell, which can be used to assess the water content of ionic resin in the proton exchange membrane or the catalyst layer [25-29]. In this paper, the operating states of each single cell in the stack under different operating conditions, such as back pressure, stoichiometry, RH and assembly pressure were analysed through the three typical DRT peaks.

3.1 DRT analysis of stack under different back pressures

Figure 3 shows the polarization curves and DRT changes of each single cell at 0/0 kPa and 80/70 kPa back pressures (anode/cathode) of the stack. Figure S2 shows the corresponding impedance data. As shown in Figure 3a, the single cell of voltage in the stack showed good consistency under no back-pressure. The average voltage and standard deviation at a current density of 500 mA/cm^2 were 0.702 V and 2.7 mV, respectively. However, when the back pressure was increased to 80/70 kPa, the average voltage and standard deviation increased to 0.751V and 3.4 mV, respectively. Combined with the DRT peak data (Figure 3b), it can be seen that, in the case of no back pressure, the internal resistance and charge transfer of each single cell in the stack were similar, whereas the corresponding P2 and P3 peaks overlapped. Meanwhile, the poor coincidence of P1 peak indicated that the water transmission state of each single cell was slightly different. However, it was not enough to cause the voltage fluctuation of the stack [30]. When the back pressure increased to 80/70 kPa, the P1 and P2 peaks changed significantly, and part of the peak began to shift towards the low-frequency direction. However, the

average voltage showed a significant increase (from 0.702 V to 0.751V), indicating that the water/gas transmission state of each single cell in the stack was chaotic [31]. On the other hand, sufficient gas supply could still be obtained in the catalyst layer of the cell [32]. At the same time, P3 peak overlapped and the intensity decreased (from 0.34 to 0.23), indicating that the wettability of the ionic resin in the proton exchange membrane or catalyst layer had improved.

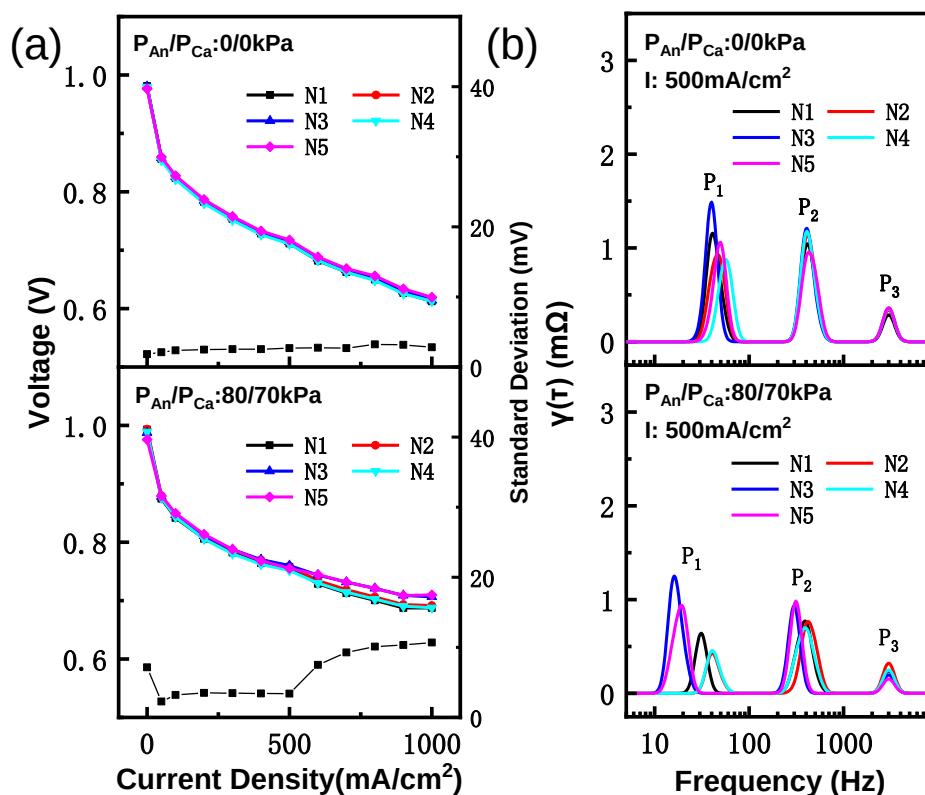


Figure 3. Polarization curves and DRT changes of each single cell in the stack when the back pressures are 0/0 kPa and 80/70 kPa (anode/cathode), respectively: (a) Polarization curves; (b) DRT data at 500 mA/cm² current density. Test condition: 40/40% relative humidity conditions, An/Ca stoichiometry is 2/2.

3.2 DRT analysis of stacks under different cathode stoichiometries

Figure 4 shows the polarization curves and DRT changes of each single cell under cathode stoichiometries of 1.6, 2 and 3. Figure S3 shows the corresponding impedance data. Because the water of the fuel cell was mainly generated at the cathode side [33-36], the stoichiometry of the cathode had a significant influence on the water transfer of the electrode [37-39]. As shown in Figure 4a, the stoichiometry led to a slight improvement in the performance of cells in the stack, with average voltages of 0.681 V (stoichiometry 1.6), 0.703 V (stoichiometry 2) and 0.712 V (stoichiometry 3). Combined with the peak data, it can be seen that, when the stoichiometry increased from 1.6 to 3, the average intensity of P1 peak decreased from 1.74 to 0.72 (peak position moved from 34.2 to 67.89), indicating that the increase in stoichiometry was conducive to the discharge of liquid water in the stack. The average

intensity of the P2 peak decreased gently from 1.23 to 0.95 (peak position moved from 389.6 to 486.3), indicating that the charge transfer in the electrode was not significantly affected. The average intensity of P3 peak increased from 0.31 to 0.42 (peak position moved from 2981.7 to 3005.8), indicating that the ohmic resistance of the cells increased slightly, which was mainly due to the decrease in the water content of the ionic resin in the membrane or the catalyst layer [40].

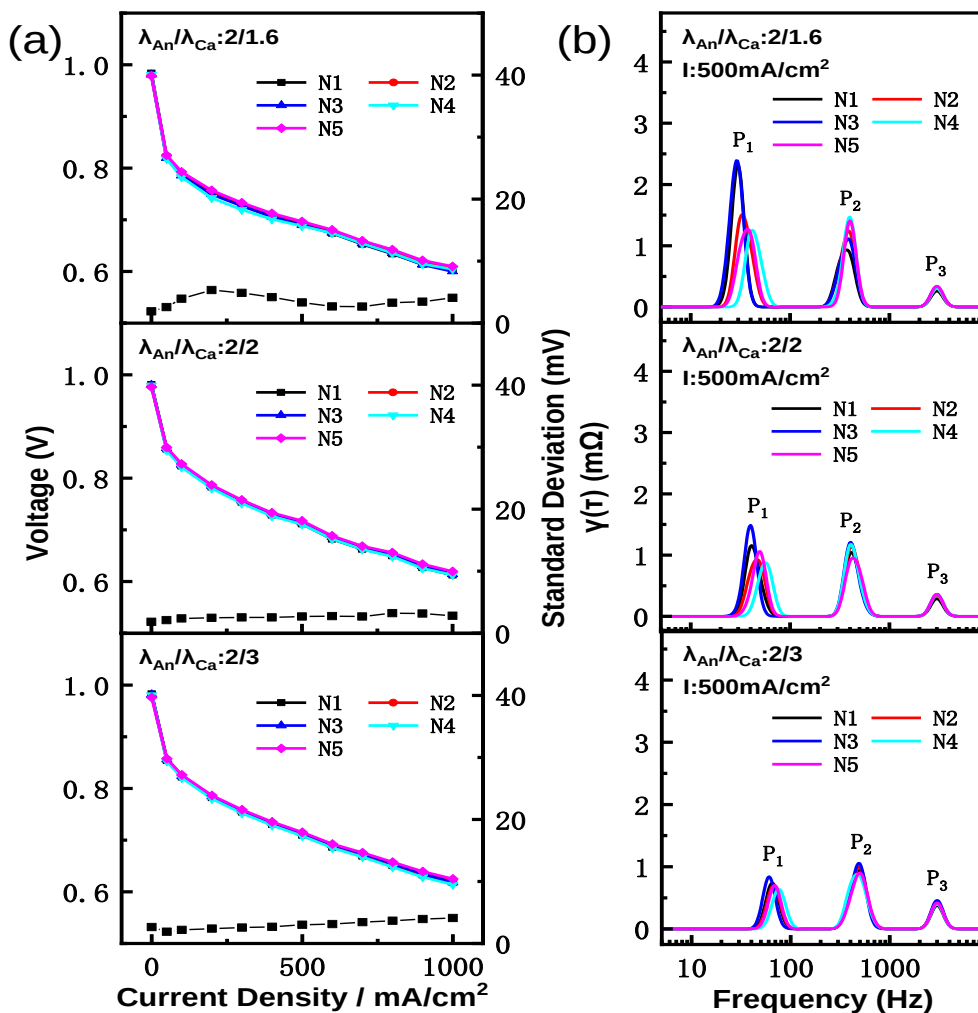


Figure 4. Polarization curves and DRT changes of each single cell in the stack when the An/Ca stoichiometry is 2/1.6, 2/2 and 2/3: (a) polarization curves; (b) DRT data at 500 mA/cm² and 1000 mA/cm² current density. Test condition: 40/40% relative humidity conditions, back pressures are 0/0 kPa (anode/cathode).

3.3 DRT analysis of stacks under different relative humidities

Figure 5 shows the polarization curves and DRT changes of each single cell of the stack under the RH values of 40/40% and 100/100%. Fig. S4 shows the corresponding impedance data. As shown in Figure 5a, the increase in RH had a great impact on the performance of the stack, and the average voltage increased from 0.702 V (RH 40/40%) to 0.724 V (RH 100/100%). Combined with the DRT peak

data (Figure 5b), it can be seen that the P_3 peak decreased from 0.34 to 0.12 with the increase in RH. This indicates that the ohmic resistance of the cells decreased significantly, which is mainly caused by the decrease in proton conduction resistance that was caused by the increase in the water content of the ionic resin in the membrane or the catalyst layer. The intensity of the P_2 peak decreased from 1.11 to 0.73, indicating that the charge transfer resistance in the electrode also decreased significantly. It is generally believed that, under higher humidification, the formation of water film and the accumulation of liquid water are likely to occur in the catalytic layer (CL) and GDL with porous media characteristics, thus preventing oxides from reaching the reactive point of reaction and increasing the reaction resistance. However, the work of Xie [41] found that under low RH, oxygen diffusion from flow field to GDL improved. Meanwhile, the degree of hydration of ionic polymer inside the CL became low, which led to the deterioration of oxygen permeability. This effect may play a dominant role in the mass transfer loss of PEMFC. Therefore, the decrease in P_2 peak value can be attributed to the increase in water content of ionic polymer resin in CL, which improves the oxygen penetration and enhances the surface charge transfer capability of the catalyst. Some previous works [42,43] also confirmed that low humidity would increase the charge transfer resistance of the cathode and reduce the electrochemically active area. There is no significant change in the intensity of P_1 peak, indicating that the increase in humidity would not cause flooding of the stack.

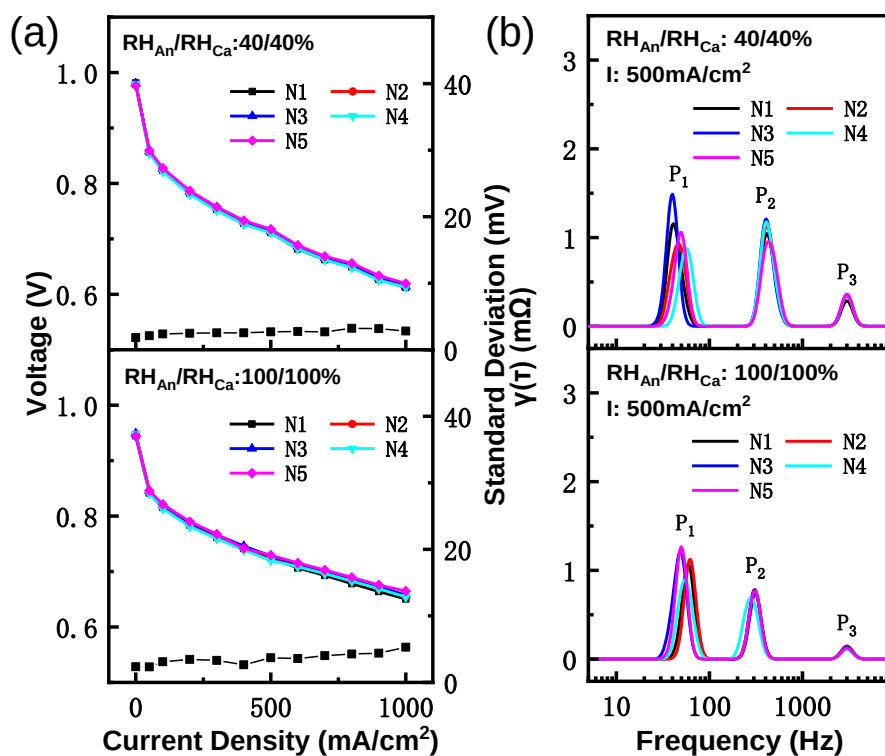


Figure 5. Polarization curves and DRT changes of each single cell in the stack under 40/40% and 100%/100% relative humidity conditions: (a) polarization curve; (b) DRT data at 500 mA/cm² current density. Test condition: An/Ca stoichiometry is 2/2, back pressures are 0/0 kPa (anode/cathode).

3.4 DRT analysis of stacks under different GDL compression ratios

Because the compressibility of the GDL is closely related to the water/gas transmission state in the electrode [44-48], this paper also tested the heap performance under different GDL compression rates. Figure 6 shows the polarization curves and DRT changes of each single cell of the stack for the GDL compression rates of 17.7%, 21.55%, 25.2% and 26.8%. Figure S5 shows the corresponding impedance data. As shown in Figure 6a, the increase in GDL compression rates had only a slight effect on the performance of the stack, with average voltages of 0.703 V (17.7%), 0.698 V (21.55%), 0.680 (25.2%) and 0.677 V (26.8%). However, the main factors affecting the performance could not be obtained from the polarization curve data. Combined with the DRT peak data (Figure 6b), the water transmission state and the internal resistance of each single cell in the stack were found to be similar. Moreover, the corresponding P_1 and P_3 peaks overlapped. Meanwhile, the P_2 peak's intensity increased gradually with the value of 1.07 (17.7%), 1.11(21.55%), 1.30 (25.2%) and 1.69 (26.8%), indicating that the charge transfer resistance in the electrode showed a significant rise. This is mainly caused by the decrease in O_2 concentration on the catalyst surface due to the lack of gas [49].

3.5 Analysis of the DRT characteristics of the stack

According to the above testing results, the voltage-time curve of each single cell at a current density of 500 mA/cm² under different working conditions and the corresponding DRT peak intensity and position are shown in Figures 7a and 7b. Figure 7c shows the operating state of the stack. Conditions 1 - 3 show the change in the cathode stoichiometries of 1.6, 2 and 3 (see Figure 5), whereas Condition 4 represents the change in the relative humidity to 100%/100% (see Figure 4). Furthermore, Condition 5 shows the change in the back pressures to 80/70 kPa (see Figure 3). Condition 6 represents the change in the back pressures to 90/80 kPa, whereas Conditions 7 - 10 represent the change in the GDL compression rates from 17.7% to 26.8% (see Figure 6). It can be seen that, under Condition 5, the voltage of the stack was the highest and also showed good consistency. However, their P_1 and P_2 peaks changed significantly, and part of the peak began to shift towards the low-frequency direction. When the back pressure was further increased to 90/80 kPa (Condition 6), the fifth single cell dropped significantly, indicating that the stack has been flooded. Therefore, it can be adjudged that, when the peak positions of P_1 and P_2 shifted to low frequency and the consistency fluctuated greatly, it indicates that the water transmission in the stack has become abnormal. Based on these results, it can be predicted in advance whether the stack is blocked with water or not. The stack shows an air supply issue for Conditions 1, 9 and 10, which was due to the reasons that the cathode stoichiometry was 1.6 and the GDL compression rate was too high. Therefore, when the peak intensities of P_1 and P_2 increased greatly, and the peak position shifted to low frequency, it indicates that the stack had the air supply issue. In all back pressure-free testing conditions (Conditions 1 - 4 and 7 - 10), the Condition 4 with relative humidity of 100%/100% exhibited the highest performance. Therefore, under other low humidification test conditions, the stack was in a water-deficient state. The intensities of P_1 , P_2 and P_3 peaks increased and the location shifted to high frequency, indicating that the stack was in a water-deficient state.

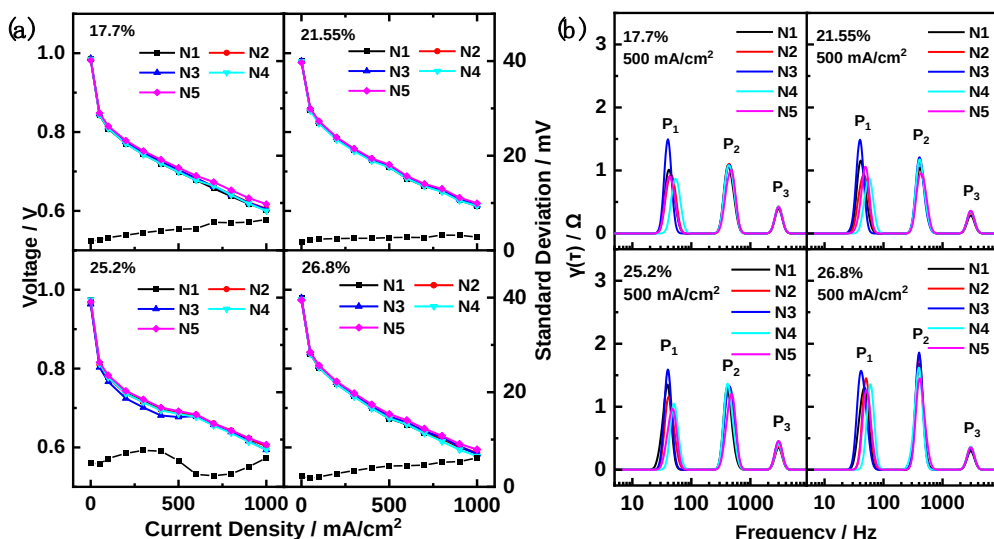


Figure 6. Polarization curves and DRT changes of each single cell in the stack when GDL compression rates are 17.7%, 21.55%, 25.2% and 26.8%, respectively: (a) polarization curve; (b) DRT data at 500 mA/cm² current density. Test condition: An/Ca stoichiometry is 2/2, back pressures are 0/0 kPa (anode/cathode), 40/40% relative humidity conditions.

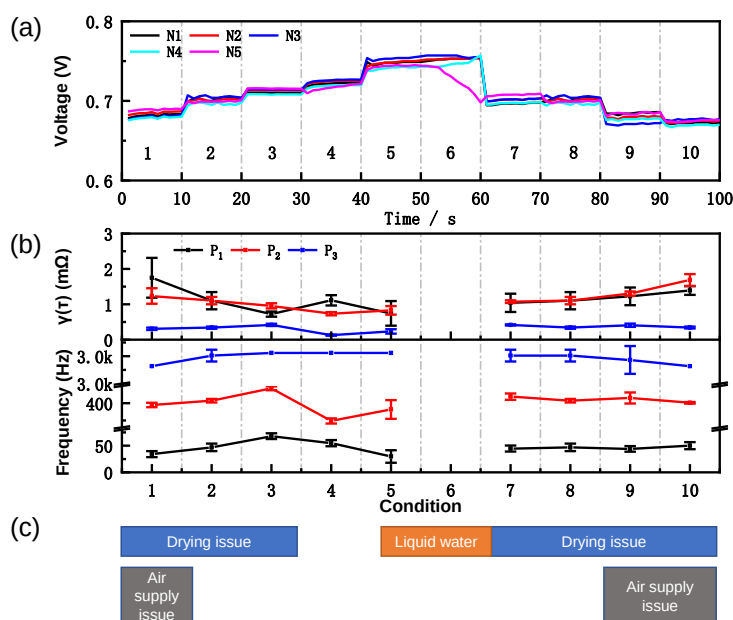


Figure 7. Voltage-time curve of each single cell at a current density of 500 mA/cm² under different working conditions (a) and their DRT peak intensity and position (b), (c) the operating state of the stack.

A previous study [50] reported that, when fuel cells were in some extreme conditions (such as dry, flooding and starvation), in addition to the changes in the intensity of characteristic peaks, their peak positions also underwent obvious displacements. However, since the experimental object is a single cell, horizontal comparison cannot be made. Therefore, the correlation between the characteristic peak's intensity and the single battery working condition was mainly studied, and the possible relationship between the change in the position of a peak and the corresponding working condition was not discussed.

In the stack, the states of each unit were objectively inconsistent. Based upon the abovementioned experimental results, it can be inferred that, when the DRT is used to study each unit in the stack, in addition to analysing the state of each unit through the characteristic peak's intensity, it is also possible to use the difference in the peak position of each unit to confirm the possible abnormal state in the stack.

4. CONCLUSIONS

In this paper, the optimal regularization parameters are obtained by analysing the distribution of relaxation times' (DRT) peaks of each cell in the stack and used to analyse the variation pattern of the operating state of each individual cell in the stack. The results show that the optimal peak can be obtained when the regularization parameter (λ) lies within the range of 10-5, whereas the change in the shape of the peak can be better used to analyse the operating state of the stack. The operating state of the stack can be adjudged and predicted by analysing the changes of P1, P2 and P3 peaks of DRT: (1) when the peak positions of P1 and P2 shift to low-frequency and the consistency fluctuates greatly, it indicates that the water transmission in the stack has become abnormal; (2) when the peak intensities of P1 and P2 increase greatly, and the peak position shifts to low-frequency, it indicates that the stack is undergoing some air supply issue; (3) when the intensities of P1, P2 and P3 peaks increase and the locations shift to high-frequency, the stack is in a water-deficient state. Therefore, by analysing the operating state of each cell in the stack through DRT, the state of the stack under conditions such as flooding, and gas shortage can be more intuitively represented. Specifically, it can predict the possible phenomenon of the stack in advance, even when no significant change has been observed in the performance of the cells, which has important practical application value.

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SUPPORTING INFORMATION

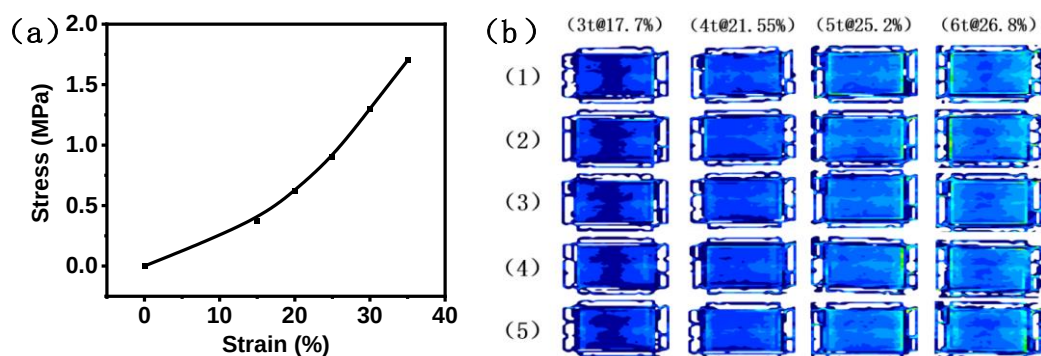


Fig. S1 (a) The stress-strain curve of the GDL; (b) the pressure distribution cloud diagram of the stack under assembly pressures of 3 t, 4 t, 5 t, and 6 t.

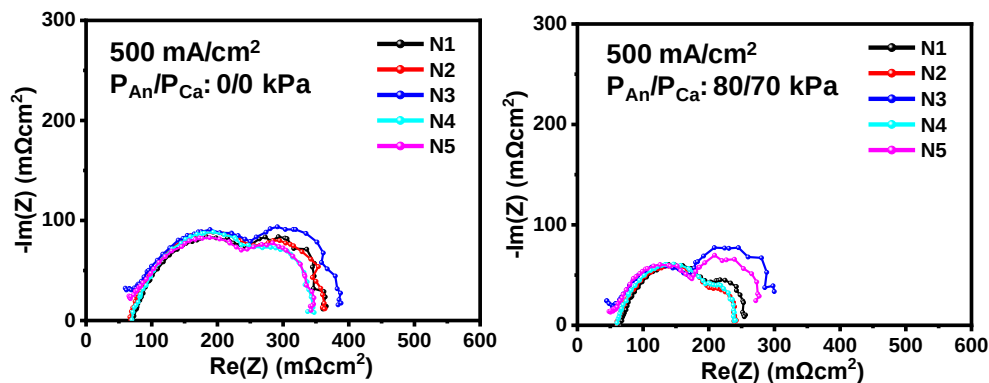


Fig.S2 Impedance data of each single cell in the stack at 500 mA/cm² current density when the back pressures are 0/0 kPa and 80/70 kPa (anode/cathode), respectively.

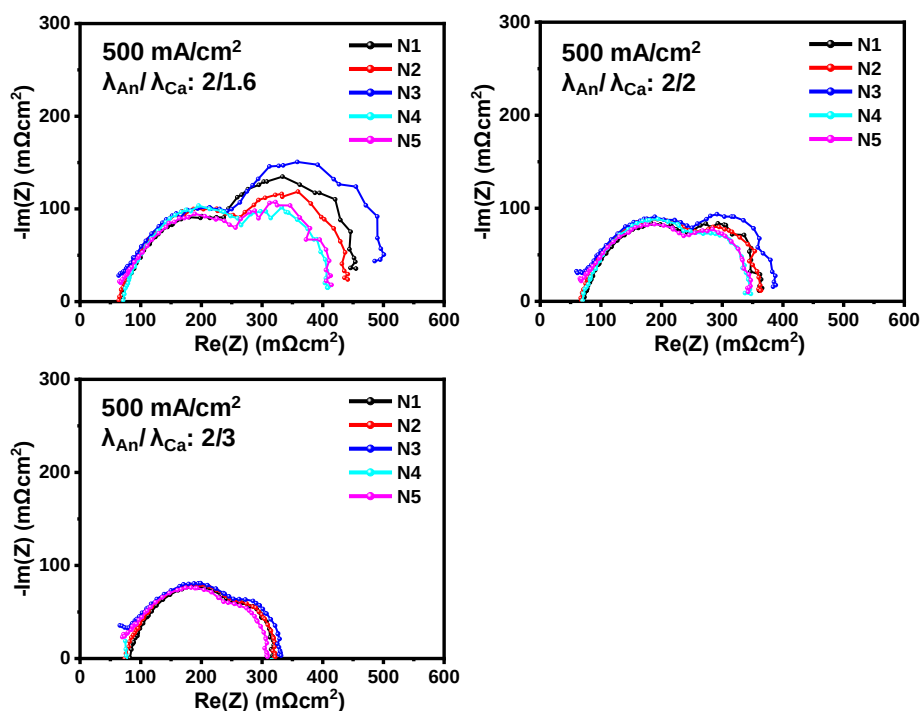


Fig. S3 Impedance data of each single cell in the stack when the An/Ca stoichiometry is 2/1.6, 2/2 and 2/3.

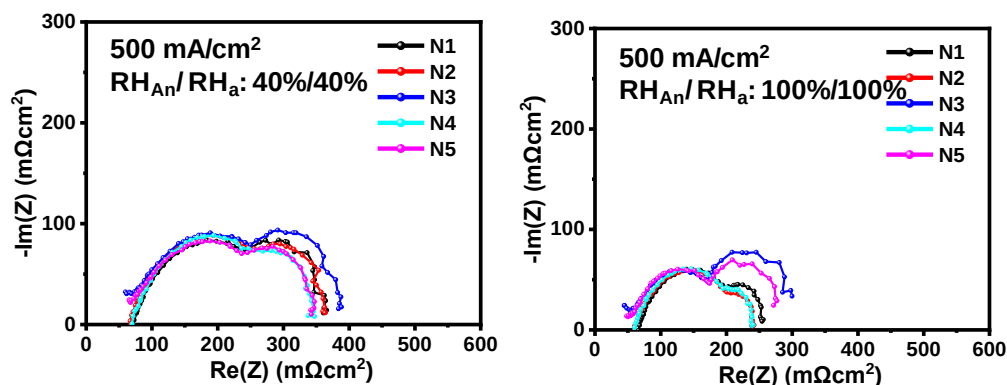


Fig.S4 Impedance data of each single cell in the stack under 40%/40% and 100%/100% relative humidity conditions.

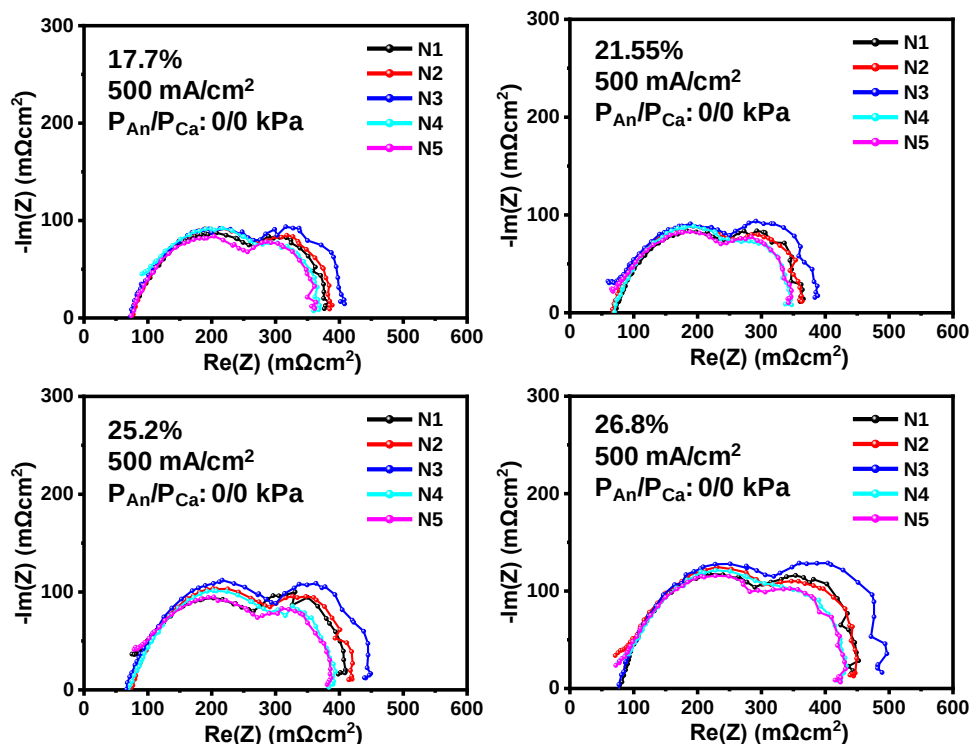


Fig.S5 Impedance data of each single cell in the stack when GDL compression rates are 17.7%, 21.55%, 25.2% and 26.8%, respectively.

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