Roll-to-roll coating methods for the manufacture of polymer electrolyte membrane fuel cell membrane electrode assemblies

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Abstract

In 2023, polymer electrolyte membrane fuel cell membrane electrode assemblies accounted for about 62% of the global fuel cell market share and this demand is expected to grow. Commercialisation requires technologies capable of cost-effective, high-volume manufacturing. The membrane electrode assembly is crucial and represents a significant cost to polymer electrolyte membrane fuel cell manufacturing. Manufacturing the electrode components for membrane electrode assemblies at high volumes can reduce production costs while delivering on the forecasted industry demands. Roll-to-roll is a mature coating technology applied in various manufacturing industries such as batteries, printed electronics, microelectronics, photovoltaics, printing, paper consumables, etc., with significant potential for polymer electrolyte membrane fuel cell manufacturing. While the high-volume production process for membrane electrode assembly manufacturing differs from laboratory-scale, the membrane electrode assembly structure remains unchanged. Roll-to-roll is a continuous coating process with speeds of 10 m/min being achieved. In this work, Microgravure™ and slot-die coating methods have been adopted and have shown promising outcomes for producing gas diffusion electrodes for membrane electrode assemblies. An overview of the Microgravure™ and slot-die coating methodologies is provided, and a discussion on preliminary results and challenges are presented. Work on Microgravure™ gas diffusion electrode manufacturing has already achieved comparable performance with spray-coated gas diffusion electrodes. The influence of coating parameters and drying on the coated catalyst layer quality is discussed.

Keywords

polymer electrolyte membrane fuel cell, gas diffusion electrode, slot-die, Microgravure™, roll-to-roll, coating window

Introduction

Fuel cells (FC) are electrochemical devices that directly and efficiently convert chemical energy into electrical energy. Polymer electrolyte membrane fuel cells (PEMFC) especially, are a crucial step towards zero carbon emissions energy utilisation and as of 2023, accounted for about 62% of the fuel cell market share (Grand View Research, 2023). However, issues related to PEMFC cost and performance are key issues that still need solving for commercialisation to be realised (Liu et al., 2023). The use of platinum (Pt) catalysts still represents 55% of the cost of PEMFC manufacturing (Liu et al., 2023), however, until alternative non-platinum group metal catalysts with performance parity to Pt catalysts are developed, alternative manufacturing processes for reducing the membrane electrode assembly (MEA) cost should be explored. One approach involves the MEA manufacturing process itself. Various methods exist to coat the MEA catalyst layers (CLs). These include paint brushing, spray-coating, doctor blade coating, gravure coating, ink-jet printing, slot-die coating, and electrospinning (Medina et al., 2023). Spray-coating is typically utilised at laboratory and small-scale production levels to coat the CL onto either the gas diffusion layer (GDL) to form the MEA or directly onto the membrane to form catalyst-coated membrane (CCM) MEAs. Although CCMs are desired due to the improved MEA performances, many challenges exist due to the swelling of the membrane when contacted with the solvents in the catalyst ink. Spray-coating is time-consuming since multiple thin layers are coated to build up the CL and require many spray coaters and operators to meet the demands of the emerging fuel cell industry. An analysis reported by Mark Debe (2012) revealed that by 2030, 10% of vehicles will be FC-powered and meeting the demand of 15 million FC vehicles per annum (each with a stack comprising 300 MEAs), production speeds of 20 m^2/min will be required. Medina and others estimated that spray coaters can coat at 0.2 m^2/min with one nozzle, thus facilities with 100 coating nozzles will be required to meet the 20 m^2/min demand (Medina et al., 2023). Continuous production processes using roll-to-roll (R2R) technologies are more suitable for meeting production targets. R2R refers to a family of manufacturing techniques in which a flexible substrate is continuously coated as it is unwound from a stock roll and transferred to a rewinding roll (Rashid et al., 2021). R2R manufacturing offers significant cost reductions and although initial capital costs can be high, these costs can often be recovered through economies of scale. R2R methods can be either self-metered or pre-metered, depending on how fluid metering is achieved. Metering is the process of controlling the rate at which fluids are applied during coating. Examples of self-metered R2R methods are gravure coating, knife, comma bar, etc. Pre-metered methods include slot-die, spray coating, extrusion die, slide curtain, etc. This work presents results from an R2R Microgravure™ method (Yasui-Seiki Mini-Labo Deluxe, Japan) and a benchtop slot-die coating method (Ossila, United Kingdom). Both techniques are suitable for high-volume production, however, various factors influence the quality of the coatings and may limit the Pt loadings required for specific PEMFC applications. Due to the many variables that need to be considered for slot-die, significant work has been dedicated to lab-scale optimisation and reported here. The challenges experienced during the practical operation of these methods are presented and discussed.

Microgravure™ overview

Gravure cylinder coating originated in the printing industry and is desirable for producing thin, uniform films at high speeds. The gravure cylinder, also called gravure roller, can typically operate in forward and reverse modes. The surface of the gravure cylinder is engraved or etched with a discrete pattern of cells that transfer the fluid from a tray to the substrate. The gravure cylinders can be engraved with various cell pattern designs such as quadrangular, tri-helical, and cells linked by small channels (Hewson et al., 2011). Figure 1 illustrates a simplified overview of the gravure coating process. Gravure coating involves the patterned cylinder partially submerged in a tray with the fluid. The gravure cylinder is rotated in the fluid and the engraved cells are filled. The doctor blade trims excess fluid from the gravure cylinder. The substrate is tensioned over a series of rollers, known as a web, and moves over the gravure cylinder. Fluid is transferred to the substrate where contact occurs, forming the coating. The cylinder design can affect the coating thickness since the grooves affect the cell opening per unit area, the average volume (cm^3/m^2), and the line count (lines per inch) (Kapur et al., 2011). Microgravure™ is a variation of gravure coating that uses a smaller diameter cylinder than regular gravure coating. The smaller diameter cylinder provides a smaller contact area and allows for a more stable coating bead to form. A stable bead is crucial for forming uniform thin coatings. Microgravure™ is a reverse kiss gravure coating method. The 'kiss' indicates that the gravure cylinder coats the substrate through a 'kissing' action and there is no backing roll to trap the substrate against the cylinder. During coating, the engraved cylinder's rotational direction is opposite to the web direction. Thus, the coating is applied to the substrate in a shearing manner (Yasui-Seiki, n.d.).

Microgravure™ coating parameters

Microgravure™ is a self-metered method and the coating thickness depends on the gravure cylinder design, cell pick-out, cylinder speed, and web speed (the substrate's speed). Cell pick-out refers to the ink transfer process from the gravure cylinder cells to the substrate, with each cylinder design having different cell pick-out rates. Microgravure™ cylinders also come with varied cell volumes

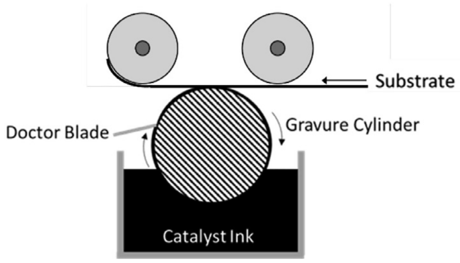

Figure 1—Simplified illustration of the Gravure/Microgravure™ coating process

similar to gravure coating. Poor pick-out leads to macroscopically defective coatings. The pick-out can be fine-tuned by precise control of the speed ratio, which is defined as the gravure cylinder speed (m/min)/web speed (m/min) (Liu et al., 2023; Park et al., 2020; Mauger et al., 2018). During coating, the fluid should be replenished at a rate equal to the consumption rate to prevent the cylinder from running dry or the fluid tray from overflowing. Both instances can affect the quality of the coating. Other parameters that affect the coating are the fluid's rheological properties (viscosity, shear thinning, shear thickening, etc.) and wettability. Fluids with contact angles < 90° are generally considered to have good wettability while fluids with contact angles > 90° have poor wettability and will be difficult to coat or result in defects. However, for coating on PEMFC gas diffusion layers (GDLs), fluids with very low contact angles may lead to excessive penetration, which may restrict gas transport through the GDL as well as impact the balance of proton and electron conductivity, leading to decreased MEA performance (Ding, Harris, 2017). Figure 2 shows photomicrographs of defective coatings obtained when (a) the catalyst ink formulation and coater web speed are not optimum and (b) the coater web speed and gravure roller speed are not optimum. In Figure 2 (a), the catalyst ink's contact angle was too high to sufficiently wet and penetrate the GDL's microporous layer, resulting in a smudged coating with an average Pt loading of 0.01 mg/cm^2. Slower web speeds may have resulted in better coating outcomes for this catalyst ink; however, slower speeds are not beneficial at production scales. A solution would be to reduce the catalyst ink's water content to lower the contact angle on the GDL's hydrophobic surface; increasing the alcohol content reduces the catalyst ink's contact angle on the GDL surface. The GDL hydrophobicity is also an important property that needs to be considered when formulating the catalyst ink. In Figure 2 (b), the coater web speed and gravure roller speed were mismatched, resulting in poor cell pick-out and a streaked coating.

Microgravure™ limitations and challenges

Microgravure™ was designed to achieve ultra-thin uniform coatings. Wet coating thicknesses between 0.8 to 80 µm can be achieved using cylinders with different engraved cell volumes. Mauger et al. (2018) used Microgravure™ to produce CLs in PEMFCs. They used a dilute catalyst ink of 3.2 wt.% catalyst and achieved Pt loadings between 0.1 - 0.13 mg/cm^2 by varying the speed ratio. Although these Pt loadings are United States Department of Energy targets for light-duty vehicle applications (U.S Department of Energy, 2023), it is insufficient for heavy-duty applications where PEMFCs will play a key role. For heavy-duty vehicle applications, Pt loadings > 0.3 mg/cm^2 are still required to meet the long durability of 30,000 hours or 1 million miles carrying heavy load (Sharma, et al., 2022). To explore the Pt loading limits of the Microgravure™ method, an 8.3 wt.% catalyst ink was coated

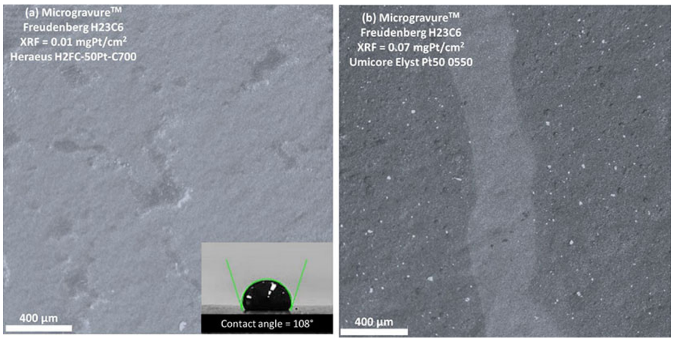

Figure 2—Defective coatings obtained with (a) a high contact angle catalyst ink and (b) incorrect speed ratio

with varying speed ratios. XRF Pt loadings of 0.164 - 0.29 mg/cm² were obtained when the speed ratio increased from 0.94–3.14. The Pt loading increased as the speed ratio increased until it reached a plateau when the gravure cylinder reached its limits. Below a speed ratio of 0.94, defective CLs were obtained, while at a speed ratio of 2.5, the Pt loading plateaued, indicating that cell pick-out was at its maximum. Increasing the speed ratio beyond this point resulted in no further increase in Pt loading. A higher catalyst wt.% ink extended the coating range to Pt loadings close to the requirements for heavy-duty PEMFC applications. Higher Pt loadings may be achieved using gravure cylinders with higher cell volumes or catalyst inks with higher catalyst wt.%, however, these may come with additional coating challenges. Since Microgravure™ is a continuous coating method, the CL is coated in a single pass compared to spray coating where multiple thin layers are coated to build the final CL. Coating the CL in a single pass results in a thick wet layer, prone to cracking during drying. Although researchers are divided on the role of cracks in the CL, eliminating cracks during single-pass CL coating remains a challenge that needs to be addressed for R2R coating methods. Figure 3 graphically illustrates how the Pt loading and CL surface crack percentage increased as the speed ratio increased. Surface cracking strongly correlates to the thickness of the catalyst layer where layers above 0.23 mgPt/cm² experienced significant surface cracking.

Figure 4 shows photomicrographs of catalyst layers corresponding to the speed ratios illustrated in Figure 3. Surface cracking can be seen to increase with increasing Pt loading. When the wet catalyst layer thickness increases, the surface layer dries faster than the inner layer, trapping solvent molecules. The only way for the trapped solvent molecules to escape is to cause stress and break through the dried surface layer, causing cracks in the surface. Adding higher boiling point solvents and crack-inhibiting additives to the catalyst inks have been explored to reduce surface cracking (Liu et al., 2024; Hasegawa, et al., 2021).

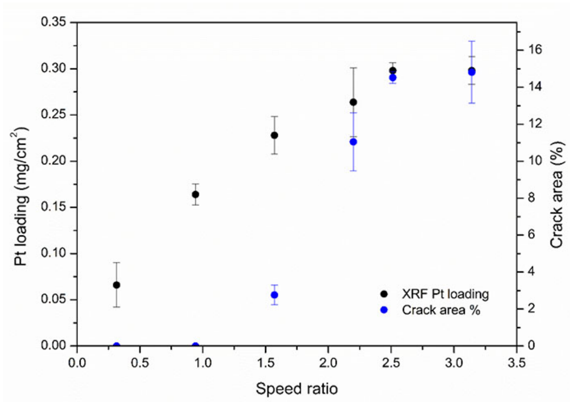

Figure 3—Microgravure™ Pt loading calibration and associated catalyst layer crack percentages

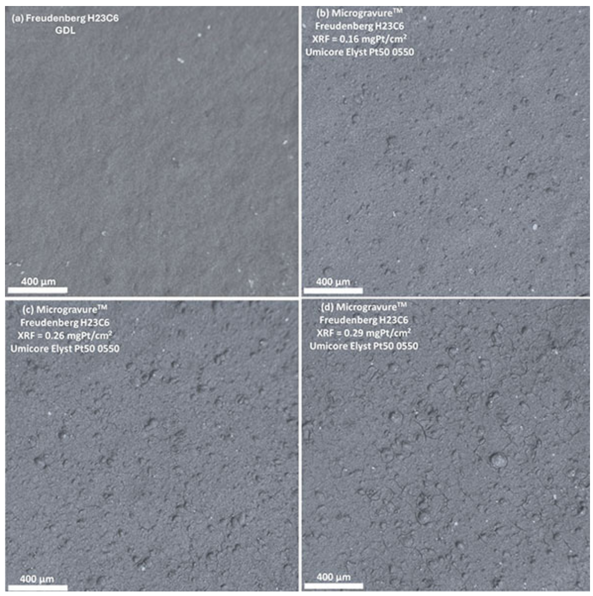

Figure 4—Microgravure™ coatings on the Freudenberg H23C6 GDL: (a) uncoated GDL, (b) coated at speed ratio = 0.94, (c) coated at speed ratio = 2.20, (d) coated at speed ratio = 2.51

Slot-die overview

The invention of slot-die is attributed to Albert E. Beguin (Sarka, Tobis, 2022). Slot-die coating is a one-dimensional technique where the substrate moves past the coating head and is coated (Vak, et al., 2016). Slot-die can be efficiently used to obtain coatings with high uniformity (Sharma et al., 2022; Creel, et al., 2022). Slot-die minimises waste by allowing precise control of the dispensed fluid. Slot-die is a pre-metered coating method consisting of a coating head and several components assembled such that the fluid enclosed in the head becomes pressurised. The fluid is forced through a slot in the head to produce uniform coatings (Vak et al., 2016). The fluid can be supplied to the head through a precision pump or compressed air. During coating, the fluid is pushed into the head and the pressure inside the head forces the fluid through a slot and onto the tensioned moving substrate. The substrate moves past the slot-die head and the fluid is continuously coated onto the substrate. After solvent evaporation and solidification of the particles, a dry uniform film on the substrate can be obtained (Ding et al., 2016). Constant fluid flow is required to prevent coating defects, thus high-precision pumps or flow meters are required to control the fluid feed. Fluctuations in the fluid flow rate can be caused by variations in viscosity, air bubbles in the system, temperature changes, and

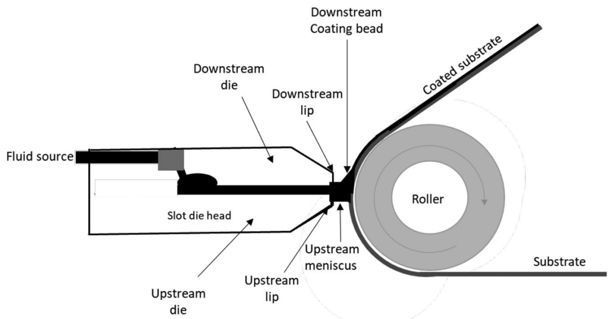

Figure 5—Illustration of the slot-die coating process

mechanical wear and tear of the flow control equipment, which will emanate as defective layers. Figure 5 shows a schematic of the slot-die coating process. Similar to gravure coating, slot-die can be coupled to roll-to-roll (R2R) processes for high-volume MEA production. Slot-die offers advantages over conventional and ultrasonic spray coating methods such as scalability, lower operational costs, and higher production rates (Sharma et al., 2022).

Slot-die coating parameters

During slot-die, the coating thickness is controlled by the gap between the slot-die-head lips and the substrate, the slot-die-lip gap, the web speed, and other factors related to the coating fluid (Ding et al., 2016; Xie et al., 2021). The relative speed between the slot-die head and the substrate translates to the coating speed (Xie, et al., 2021). The coating speed, gap height, ink rheology (viscosity, shear thinning, etc.), substrate properties (roughness, hydrophobicity, hydrophilicity, etc.), and drying conditions can influence the quality of the obtained coatings. If proper control of these conditions is not met, defects such as ribbing, barring, rivulets, and air entrainment can occur (Ding, Harris, 2017; Ding et al., 2016). Figure 6 shows examples of coating defects that occurred when the optimum coating conditions were not met. The wet coating thickness (t_{wet}) in traditional slot-die coating can vary from ten to a few hundred microns. With a tensioned web, a wet thickness of less than 5 microns can be obtained (Ding et al., 2016). Wet thickness can be calculated using Equation 1 where v is the coating speed (cm/s), Q is the fluid dispense rate (cm^3/s) and w is the coating width (cm). Stähler et al. (2019) used a similar equation to determine the wet thickness. Determining t_{wet} is crucial for setting the gap height to prevent the gap from being too small, causing scraping and pooling of the coated layer or too large, affecting the formation of a stable coating bead and leading to defects.

$$t_{wet} = \frac{v}{Q \times w} \qquad [1]$$

Determining operating limits for slot-die is a complex process where various factors and competing forces are taken into account, as discussed elsewhere (Creel et al., 2022; Ding, et al., 2016). Figure 7 shows coating windows developed for three catalyst inks utilising the same 50 wt.% Pt/C catalyst but different ionomer-to-carbon (I/C) and water/alcohol ratios. The coating window is a stable operational window where uniform and defect-free coatings are possible (Chang et al., 2007). Contrary to Newtonian fluids, fluids such as colloidal suspensions and polymeric solutions can have very complex rheological properties such as shear thinning, extension thickening, and viscoelasticity, which can affect the force balance in the coating bead and the subsequent operation limit of the coating process (Ding et al., 2016). The catalyst ink for MEA manufacturing is a colloidal suspension consisting of particles and

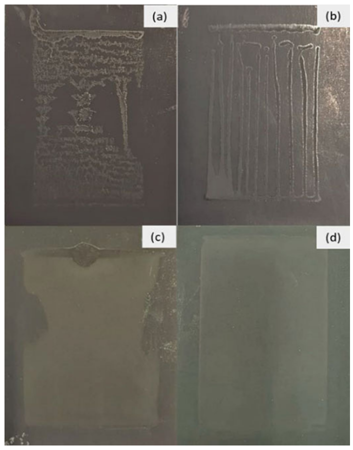

Figure 6—Coating qualities obtained for slot-die coated catalysts layers (a) barring, (b) ribbing, (c) unstable meniscus and coating bead and (d) defect-free coating

ionomers, sometimes with polymer additives that typically exhibit shear thinning when shearing is applied. However, the rheological properties are strongly influenced by the composition of the ink, such as catalyst type (support type and metal %), ionomer content, and solvent type. A simple viscocapillary model as described by Creel et al. (2022), Koh et al. (2012), Higgens and Scriven (1980), and Lee et al. (2011) was adopted to develop the coating windows. The parameter inputs for this model were the viscosity of the ink (N/m^2.s), surface tension (N/m), coating width (m), gap height (m), upstream lip width (m), downstream lip width (m), and static and dynamic contact angles. From Figure 7 it is observed that increasing the I/C ratio from 0.9 (black coating window) to 1.8 (red coating window) resulted in a slight broadening and position shifting of the coating window. Higher ink flow rates were required at the same coating speed for the 1.8 I/C ratio catalyst ink to obtain defect-free coatings. The coating windows for the catalyst ink with a water/alcohol ratio of 9 (green coating window) had a very narrow coating window due to the low wettability of this ink. The high water content of the catalyst ink caused a contact angle of 120° on the GDL. Although a narrow coating window was produced, Pt loadings in this coating window were limited to ~0.1 mg/cm^2, a loading too restrictive for MEA manufacturing. It should be noted that coating windows are strongly influenced by the GDL type as the surface tension will change.

Slot-die limitations and challenges

Slot-die coating equipment generally requires a higher upfront investment cost than simpler coating methods. Slot-die coating is also more complex and requires precise control of coating variables, which are often obtained through coating window development. Fluids have different coating windows depending on the fluid makeup. Colloidal suspensions and polymeric solutions will have different coating windows to Newtonian fluids. Figure 7 demonstrates how the I/C ratio and water/alcohol ratio influenced the coating windows of catalyst inks. As with Microgravure™, coating catalyst layers in a single pass creates drying issues resulting in surface cracks. The crack area increases with the coated layer thickness required to achieve Pt loading targets in heavy-duty PEMFC. Uniformity of the coated layer is easily affected by deviations in the coating parameters, thus high-precision equipment is crucial. Figure 8 compares the catalyst layers obtained through slot-die and spray-coating utilising two commercial catalysts, i.e. the Umicore Elyst Pt50 0550 (carbon black support) and the

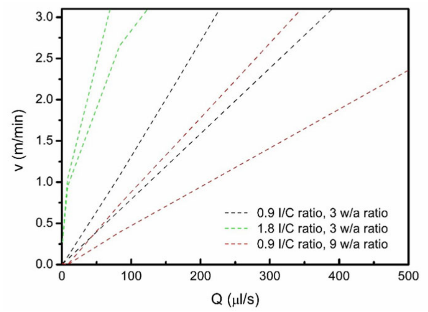

Figure 7—Coating windows for catalyst inks with different I/C and water/alcohol ratios

Heraeus H2FC-50Pt-C700 (high surface area carbon support). For both catalysts, the spray coated counterpart resulted in uniform catalyst layers with no visible cracks, since spray coating is a multi-layered process and drying occurs continuously throughout the coating process, thereby mitigating crack formation. The slot-die catalyst layers were deposited as a single layer and thus cracked during drying. The drying process of the wet coating significantly influences the morphology and microstructure of the catalyst layers. Solvent evaporation, sedimentation, agglomeration, assembly, packing, and internal particle migration due to concentration and temperature gradients, are the dominating factors that influence catalyst layer microstructure formation (Liu et al., 2023). Cracks have been reported to have a detrimental effect on PEMFC performance and have been linked to pin-hole formation in the membrane, local flooding, catalyst erosion, free radical generation, and increased catalyst layer resistance (Liu et al., 2024). Optimising the catalyst layer microstructure is crucial for PEMFC performance and is an important challenge that needs to be addressed in slot-die coating.

Influence of Microgravure™ and slot-die coating on MEA performance

Beginning-of-life performances of MEAs fabricated using the Microgravure™, slot-die, and spray-coated GDEs were evaluated. Figure 9 (a) shows the polarisation curves of MEAs utilising the Umicore Elyst Pt50 0550 catalyst. The MEAs composed of the Microgravure™ and spray-coated GDEs showed comparable performance with the Microgravure™ having marginally better performance at 0.6V. However, at higher current densities, the spray-coated MEA showed improved performance, achieving higher peak power density. The improvement at higher current densities for the spray-coated MEA can be linked to the microstructure of the catalyst layer with fine cracks and craters in the Microgravure™ catalyst layers, possibly leading to localised flooding and higher mass transport resistance. The performance metrics are summarised in Table 1. The slot-die coated MEA showed notably lower performance than the Microgravure™ and spray-coated MEAs, which could be due to the highly cracked surface of the catalyst layers. Although the three MEAs had similar electrochemical surface areas (ECSA), the spray-coated MEA showed significantly better ORR activity indicating better Pt utilisation. Figure 9 (b) shows the polarisation curves of MEAs utilising the Heraeus H2FC-50Pt-C700 catalyst. Since the ink containing this catalyst did not coat with Microgravure™, no MEA result was available. Microgravure™ coating of this catalyst will be explored in future work. The figure compares the performance of the slot-die MEA and the spray-coated MEA. As observed with the Umicore catalyst, the spray-coated MEA outperformed the slot-die MEA. In this

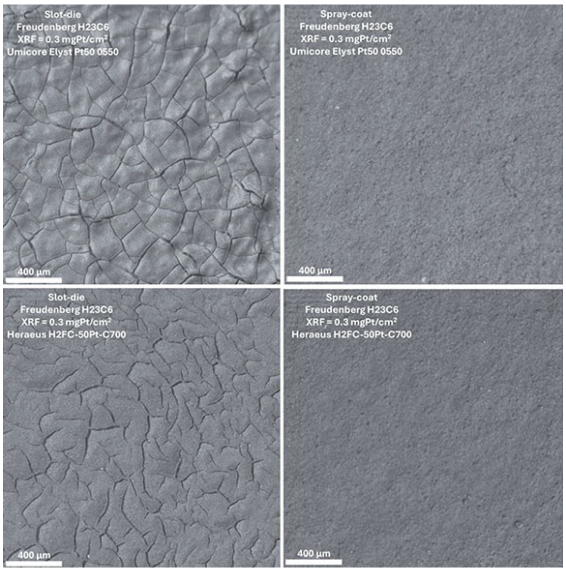

Figure 8—Micrographs of slot-die and spray-coated GDEs. Coatings with Umicore and Heraeus catalysts utilising carbon black and a high surface area carbon, respectively, are also compared

case, the spray-coated MEA exhibited a larger ECSA of 102.69 m^2/gPt compared to 71.39 m^2/gPt, demonstrating that the uniform catalyst layer in spray-coating resulted in better Pt utilisation. Optimising drying conditions, especially for slot-die, is crucial for uniform crack-free catalyst layers and improved MEA performance. Although the spray-coated MEA with Umicore catalyst achieved the highest peak power density, the Microgravure™ MEA with Umicore catalyst and the spray-coated MEA with Heraeus catalyst achieved comparable peak power densities at higher voltages, indicating less Pt degradation should occur during peak PEMFC operation.

Conclusions

Microgravure™ and slot-die coating methods were evaluated for GDE fabrication, and preliminary MEA performance results indicate that Microgravure™ has a strong potential for high-volume GDE production. Slot-die coated catalyst layers yielded significant cracking during drying, affecting the MEA performance. Although Microgravure catalyst layers exhibited cracking, it did so to a much lesser extent than slot-die coating. Addressing challenges related to catalyst layer drying is crucial to producing high-performance MEAs at high-volume scales. The use of catalyst inks with higher boiling point solvents and/or crack-inhibiting additives will be investigated in future work. Moreover, optimising other catalyst ink variables, such as the solvent composition, is crucial in Microgravure™ and slot-die methods to ensure wettability and catalyst ink spreading on the GDL surface. Catalyst inks with high contact angles do not wet the GDL surface well and are prone to producing defective coatings. In slot-die, precise control of other coating variables such as web speed, ink dispensing rate, gap height, etc. is also crucial since deviations in any of these variables will lead to defective coatings. The Microgravure™ method is generally simpler than slot-die, requiring control of fewer parameters. However, Microgravure™ is restricted by a wet layer thickness maximum, which limits the scope of application of the method.

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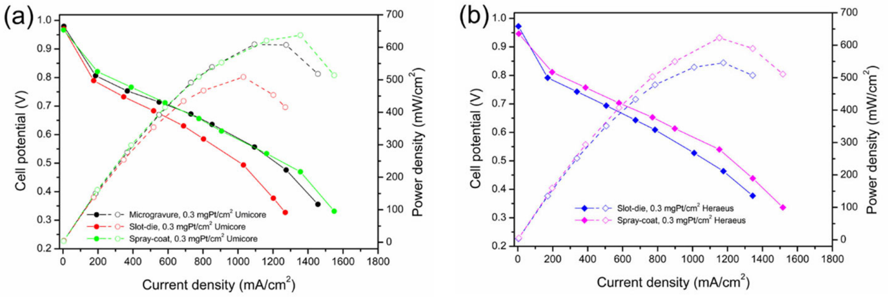

Figure 9—Polarisation and power curves of Microgravure™, slot-die, and spray-coated MEA with (a) Umicore and (b) Heraeus catalysts. Cell temperature = 80°C, a/c: RH = 100/80, 2/2 bar backpressure, hydrogen/air flow rates = 0.43/2 nlpm. Cell fixture pressure at 4.8 bar

Table 1

Electrochemical properties of the MEAs evaluated

MEA name	Current density @ 0.6V (mA/cm²)	Power density @ 0.6V (mW/cm2)	Peak power density (mW/cm²)	ECSA (m²/gPt)	IR corrected MA @ 0.9V (mA/mgPt)	IR corrected SA (µA/cm² Pt)
MG - Umicore	961	571	609 @ 0.56V	63.54	34.05	59.6
SD – Umicore	765	456	509 @ 0.49V	59.11	44.54	75.34
SC – Umicore	946	564	637 @ 0.47V	61.24	85.22	139.16
SD – Heraeus	810	484	546 @ 0.46V	71.39	23.9	41.04
SC – Heraeus	945	563	623 @ 0.54V	102.69	59.43	57.87

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