Roll-to-roll coating methods for the manufacture of PEMFC MEAs

Cecil Felix
Olivia Barron

08 April 2025

Presentation outline

1. Introduction
2. Microgravure™ overview
3. Microgravure™ coating parameters
4. Microgravure™ limitations and challenges
5. Slot-die overview
6. Slot-die coating parameters
7. Slot-die limitations and challenges
8. Microgravure™ vs. slot-die vs. spray coat
9. Conclusions and future work
10. Acknowledgements

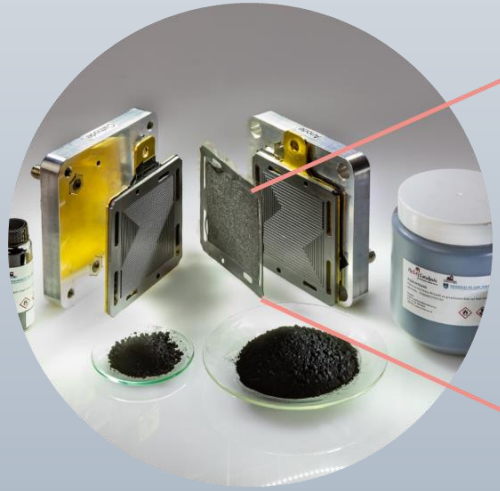
Polymer electrolyte membrane fuel cells (PEMFC)

PEMFC

- Efficient H_2 conversion into clean electricity
- Crucial in hydrogen economy

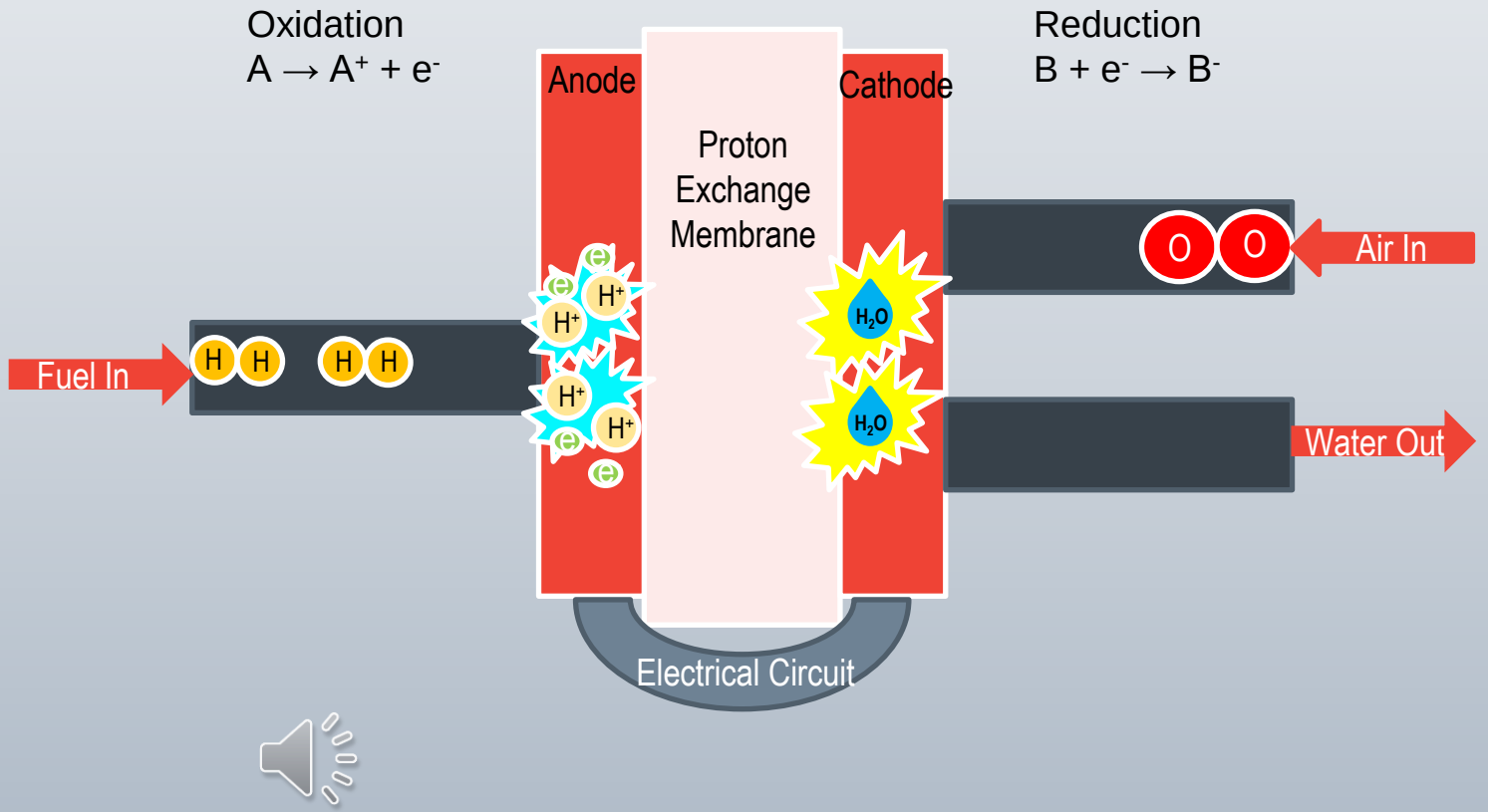
MEA

- Core of PEMFC
- HOR and ORR takes places
- High-volume production for cost reduction



Membrane electrode assembly (MEA)

1. Introduction (Cont.)



1. Introduction (cont.)

Roll-to-roll (R2R) technology

- **Mature technology used in many industries**
- **Continuous processing to meet MEA production targets**
- **Significant cost reductions → high initial capital but recovered through economies of scale**
- **Couples to self-metered methods → gravure, knife, comma bar, etc.**
- **Couples to pre-metered methods → slot-die, spray coating, extrusion-die, etc.**

Ultrasonic spray coater

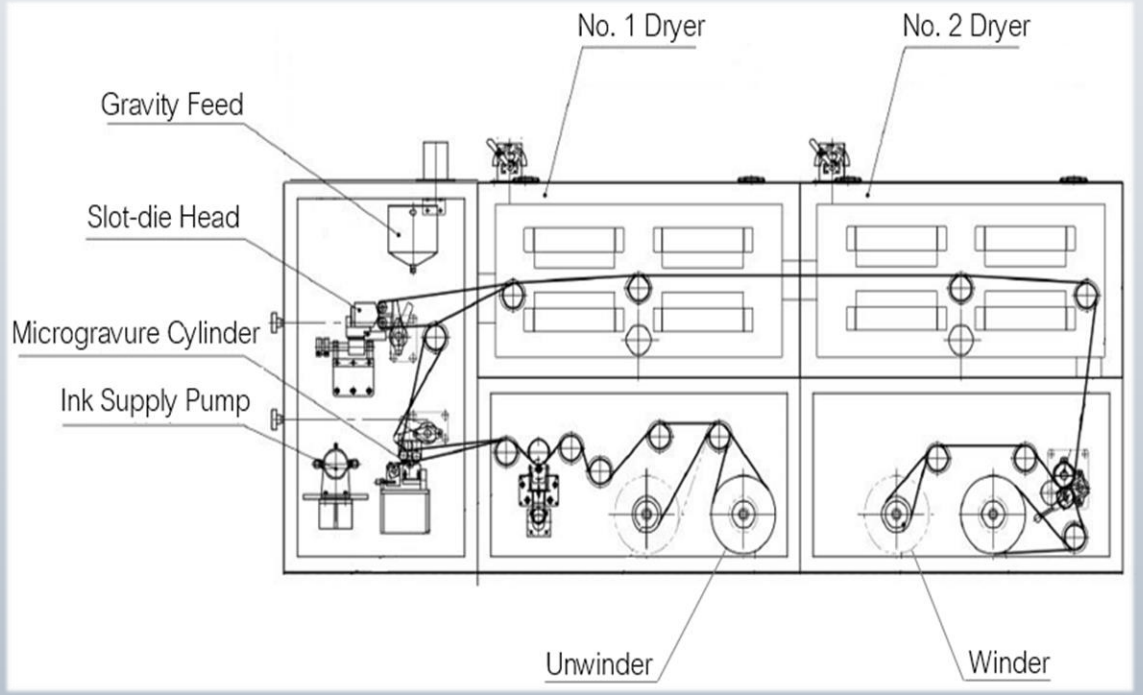
Lab to pilot-scale


R2R coater

1. Introduction (Cont.)

Roll-to-roll overview

- **R2R equipment consists of many components**
- **Coating speed of 10 m/min have been reported**
- **Spray coaters typical 0.2 m^2/min**
- **Pilot coater 5 Lm/min or 1.5 m^2/min**
- **Mintek has Microgravure™ and slot-die coating capabilities.**



1. Introduction (Cont.)

Mintek's MEA production process

Catalyst ink development and QC

- Catalyst ink formulations
- High shear mixing
- Microfluidising
- Particle size analysis
- Rheology

GDE/CCM fabrication

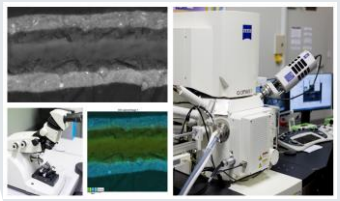
- R&D coatings
- R2R slot-die
- R2R Microgravure™
- R2R calendering

GDE/CCM characterisation

- X-ray fluorescence
- Optical microscopy

MEA assembly and evaluation

- GDE cutting
- Membrane cutting
- MEA hot-pressing
- Single cell testing
- MEA gasketing*
- Stack testing*



MEA characterisation

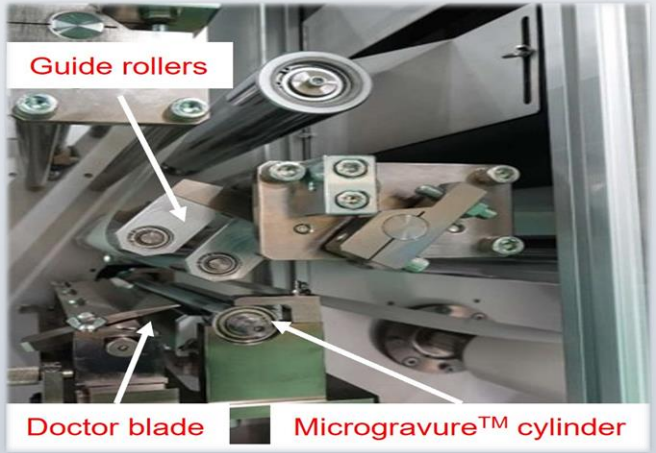
- Sample preparation
- Ultramicrotomy⁺
- High-res SEM⁺
- EDS⁺

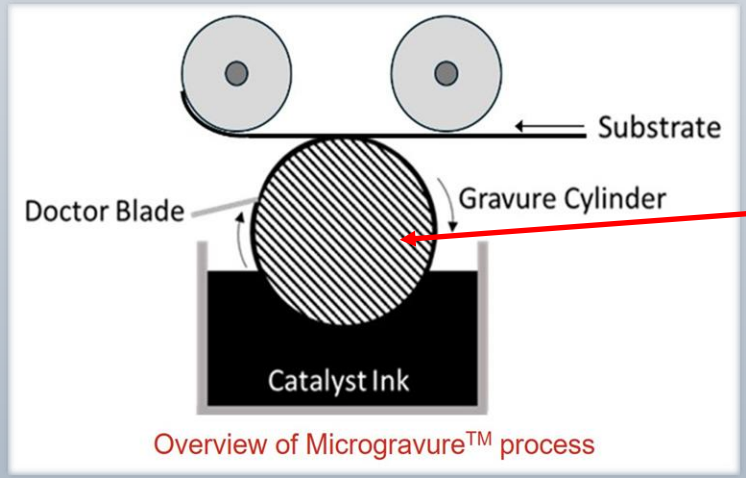
* Future plans

⁺ Outsourced

2. Micrograv ure™ overview

- **Gravure coating originated in the printing industry**
- **Micrograv ure™ – variation of gravure coating**
- **Self-metered method for high-speed thin coatings**
- **Uses cylinders with discreet patterned cells – quadrangular, tri-helical, channel-link cells → different cell volumes**





Overview of Micrograv ure™ process

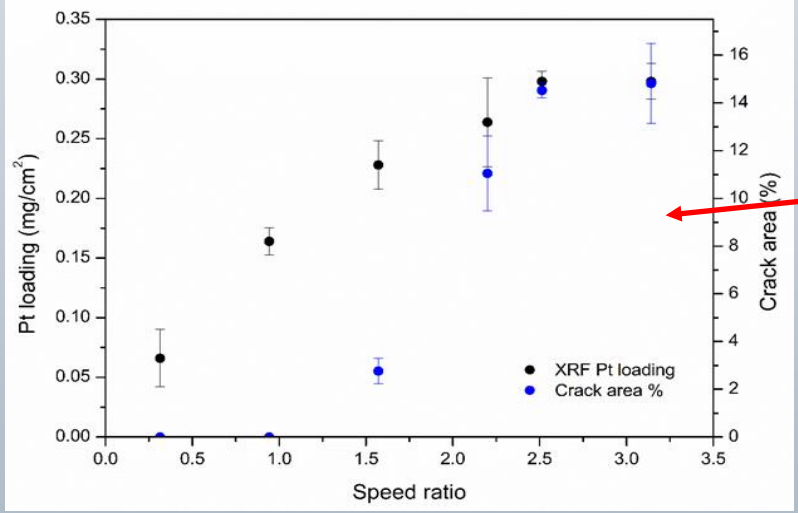

R30 mesh tri-helical cylinder

3. Microgravure™ coating parameters

- **Coating thickness depends on cylinder design, cylinder speed, cell pick-out (ink transfer) and web speed**
- **Cell pick-out can be fine-tuned by the speed ratio (cylinder speed/web speed)**

Mesh Lines/inch	Wet Coat Thickness μ *	Mesh Lines/inch	Wet Coat Thickness μ *
25	50 - 80	85	13 - 22
30	30 - 45	90	8 - 16
36	28 - 43	95	7 - 15
38	25 - 40	100	6 - 14
45	28 - 43	110	6 - 13
50	25 - 35	120	5 - 11
55	20 - 30	150	4 - 9
60	21 - 31	180	3 - 8
65	13 - 22	200	2 - 5
70	16 - 30	230	1.5 - 3.5
75	20 - 30	250	0.8 - 2
80	12 - 20		

Microgravure™ cylinder meshes and coating thicknesses



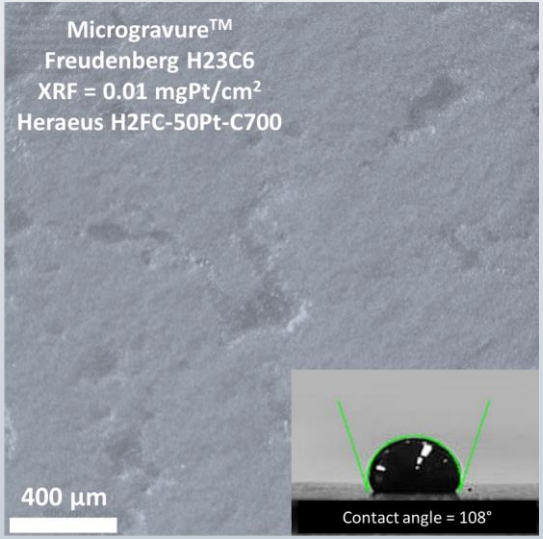
Pt loading calibration with R30 mesh cylinder

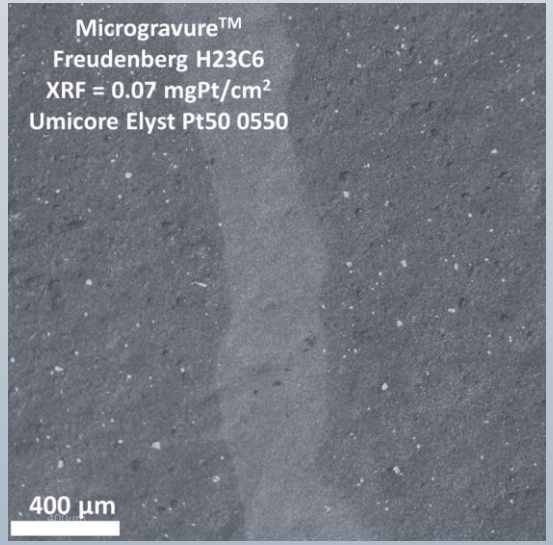
- **Calibration predicts Pt loadings**
- **Pt loading increases with speed ratio**
- **Catalyst layer cracking increases with Pt loading**
- **Beyond a certain speed ratio, Pt loading plateaus (max. cell pick-out)**

3. Microgravure™ coating parameters (Cont.)

Defects

- **Catalyst ink contact angle important for coating (<90°)**
- **High contact angle lead to defective coating**
- **Catalyst ink should sufficiently wet the hydrophobic GDL**
- **Coating parameters different across various GDLs**





- **Incorrect speed ratio**
- **Low-speed ratio → insufficient cell pick-out (ink transfer)**

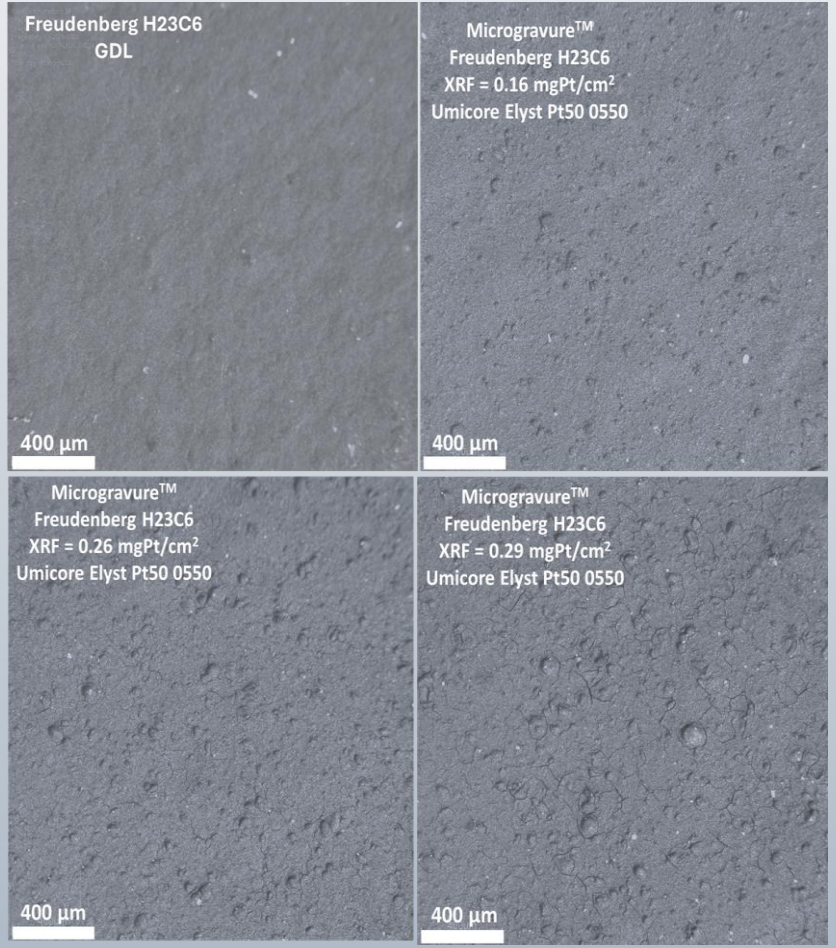
4. Micrograv ure™ limitations and challenges

Limitations

- **Wet coating range is 0.8 - 80µm**
- **Max. viscosity 200 mPa.s**
- **USDOE targets – 0.125 and 0.35 mgPt/cm² for light- and heavy-duty PEMFC**
- **We achieved max. ~0.3 mgPt/cm²**

Challenges

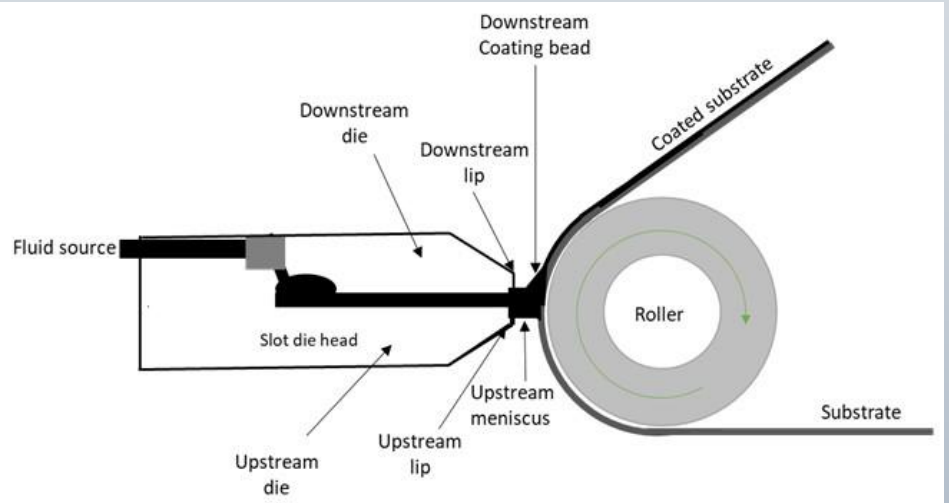
- **Catalyst layer cracking more pronounced with increasing Pt loading**
- **Cracking impacts MEA performance, especially durability**



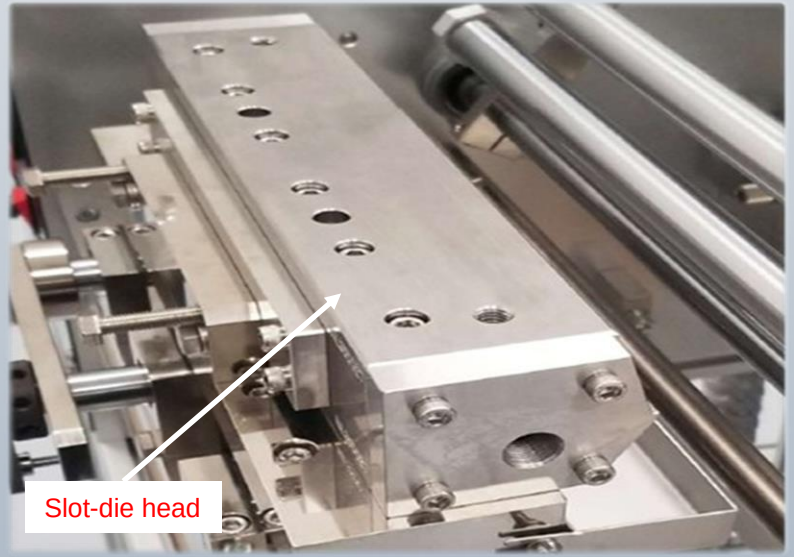
MicrogravureTM coatings on the Freudenberg H23C6 GDL at different speed ratios

5. Slot-die overview

- **Pre-metered method → allows precise ink dispensing and minimises waste**
- **Consists of a head with a slot for dispensing ink onto the substrate**
- **Pump or compressed air feeds and pressurises ink**
- **<u>Advantages</u> → scalable, low operational costs and high production rates**



Overview of slot-die process



6. Slot-die coating parameters

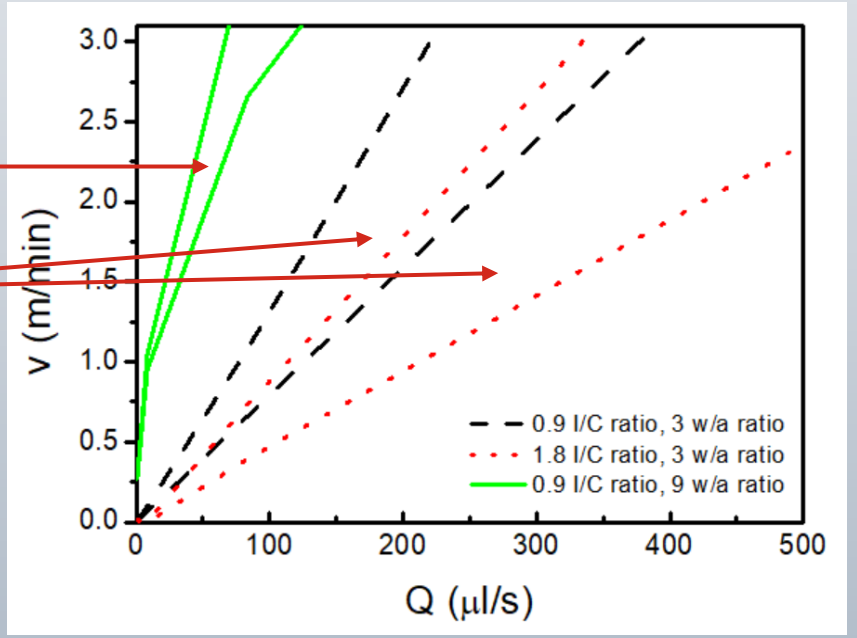
- **Coating thickness controlled by gap height, lip gap, web speed and ink properties**
- **Coating window → slot-die coating predictions**

High-water ink – narrow coating window with low Pt loading

Ionomer-to-carbon shift coating window

0.15 – 0.9 mgPt/cm^2 achieved

Coating speed up to 3 m/min achieved

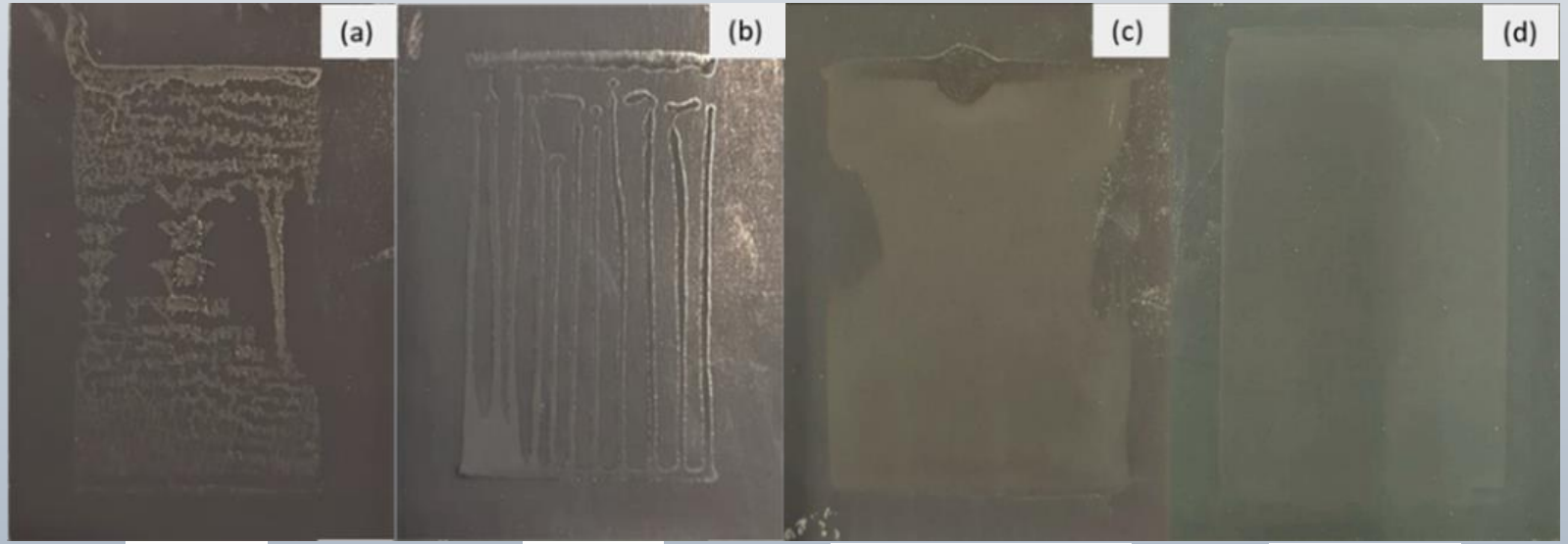


Coating windows developed with simple viscocapillary model

Slot-die coating parameters (Cont.)

Defects

- **Common defects – ribbing, barring, air entrainment and rivulets**
- **Good meniscus and stable coating bead crucial for slot-die coating**



(a) barring (b) ribbing (c) Unstable coating bead (d) Defect -free

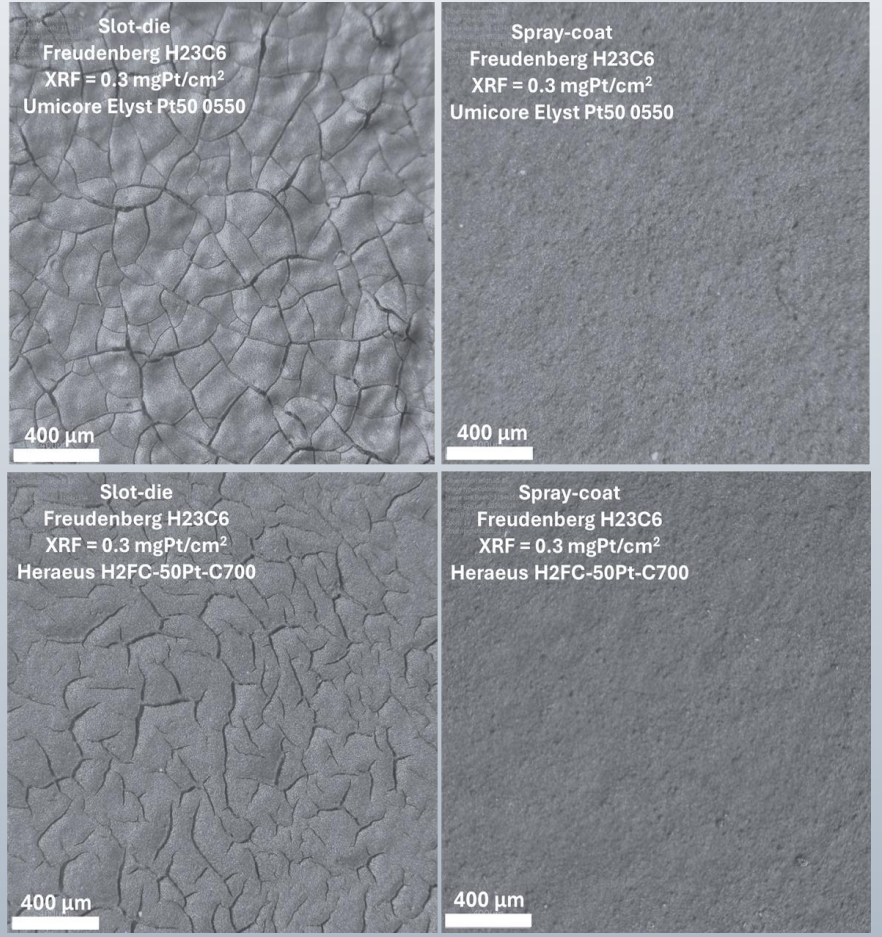
7. Slot-die limitations and challenges

Limitations

- **Min. wet coating thickness 20 µm**
- **Min. viscosity 100 mPa.s**

Challenges

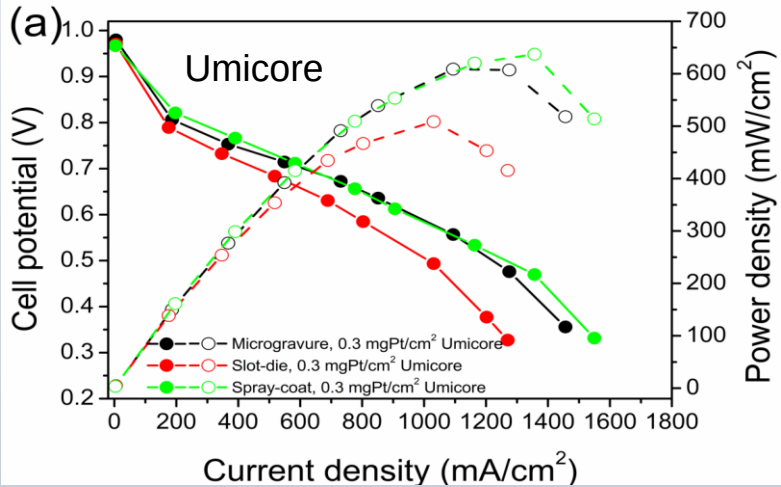
- **Coating parameters are complex to control**
- **Parameters differ across catalyst inks and GDLs**
- **Precise and stable fluid feed required to avoid defects**
- **Catalyst layer cracking is significant cf. spray-coating**

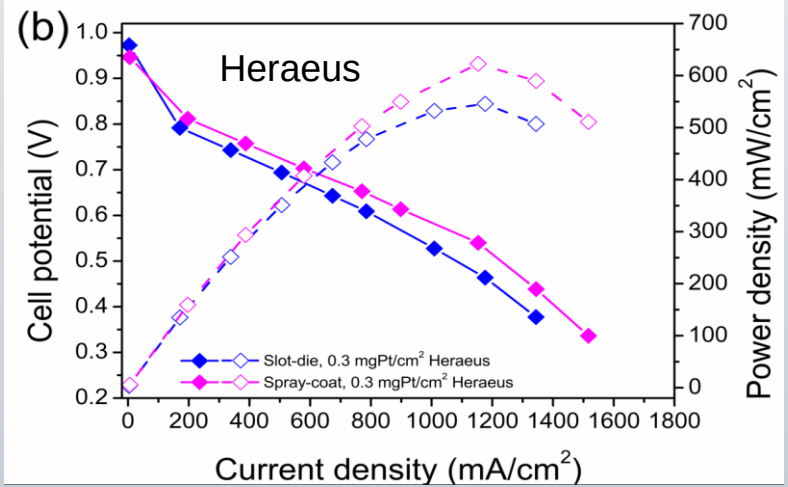


Slot-die coatings on the Freudenberg H23C6 GDL with different Pt/C catalysts

8. Microgravure™ vs. slot-die vs. spray coat

MEA results





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

MG = Microgravure™, SD = slot-die, SC = spray-coated

9. Conclusion and future work

Conclusions

- **Slot-die coating more complex than Microgravure™**
- **Slot-die has a wider Pt loading range, achieving light- and heavy-duty PEMFC Pt targets**
- **Preliminary MEA testing shows Microgravure™ performance compares to spray-coat MEAs**
- **Microgravure™ and slot-die catalyst layers crack with the latter to a greater extent**

Future work

- **Solve the issue of catalyst layer cracking**
- **Direct-to-membrane coatings for CCM development**
- **Achieve higher coating speeds**

10. Acknowledgements

1. Department of Mineral Resources & Energy
2. HySA Catalysis (Mintek)

mineral resources & energy
Department:
Mineral Resources and Energy
REPUBLIC OF SOUTH AFRICA

HySA Catalysis
Hydrogen South Africa

Thank you for Listening

200 Malibongwe Drive
Randburg
South Africa

Tel: +27 11 709 4111

www.mintek.co.za

Private Bag X3015
Randburg
2125
South Africa

Mintek MintekSA Mintek_RSA Mintek_RSA