Karlsruhe Institute of Technology

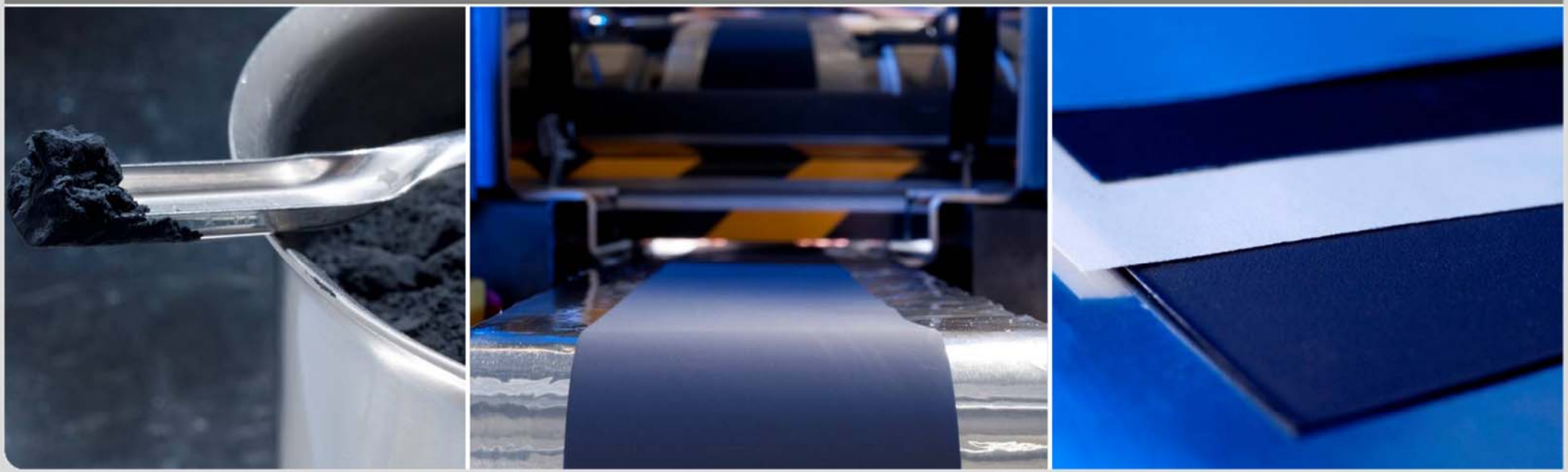
Institute for Applied Materials

Processing of water-based electrode pastes for lithium nickel manganese cobalt oxide (NMC) batteries

F. A. Çetinel, D. Nötzel and W. Bauer

Shaping 5, 29-31 January 2013, Mons, Belgium

Institute for Applied Materials – Material Process Technology (IAM-WPT)



Why using water-based pastes?

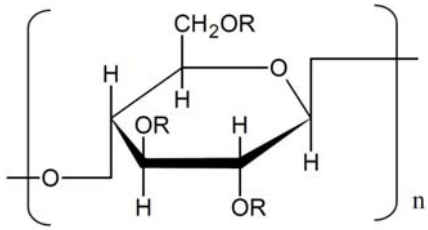
- **NMP (N-Methyl-2-pyrrolidon)** is widely and successfully used as an organic solvent for paste formulations applied for manufacturing of electrodes
  - Toxic
  - Irritating
  - Teratogenic
  - Flammable
  - Expensive (30-50 €/l)
  - High efforts and costs for operational safety, explosion protection, waste gas treatment, waste management

Issues of water-based processing

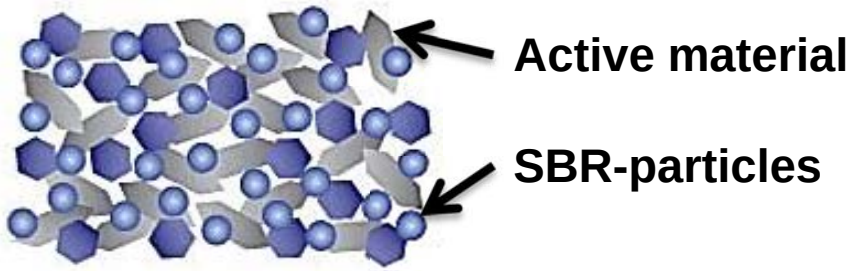
- Possible degradation of active materials
- Poor paste homogeneity due to tendency to agglomeration
- Increasingly often occurrence of drying cracks
- Narrow process range for paste preparation and coating
- *Currently, water-based processing exhibits state-of-the-art for industrial fabrication of anode electrodes*
- *Almost no industrial application of water-based pastes for cathode electrodes fabrication*

Additives for water-based pastes

- Na-Carboxymethylcellulose (CMC)
  Thickening, dispersing and adhesive agent



- Rubber or latex polymers
  e.g. Styrene-Butadiene-Rubber (SBR)
  Binder, adhesive agent



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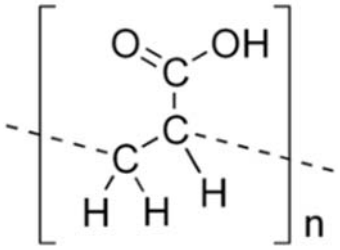
- Polyacrylic acid (PAA)
  Dispersing agent



- …

Preparation of water-based pastes

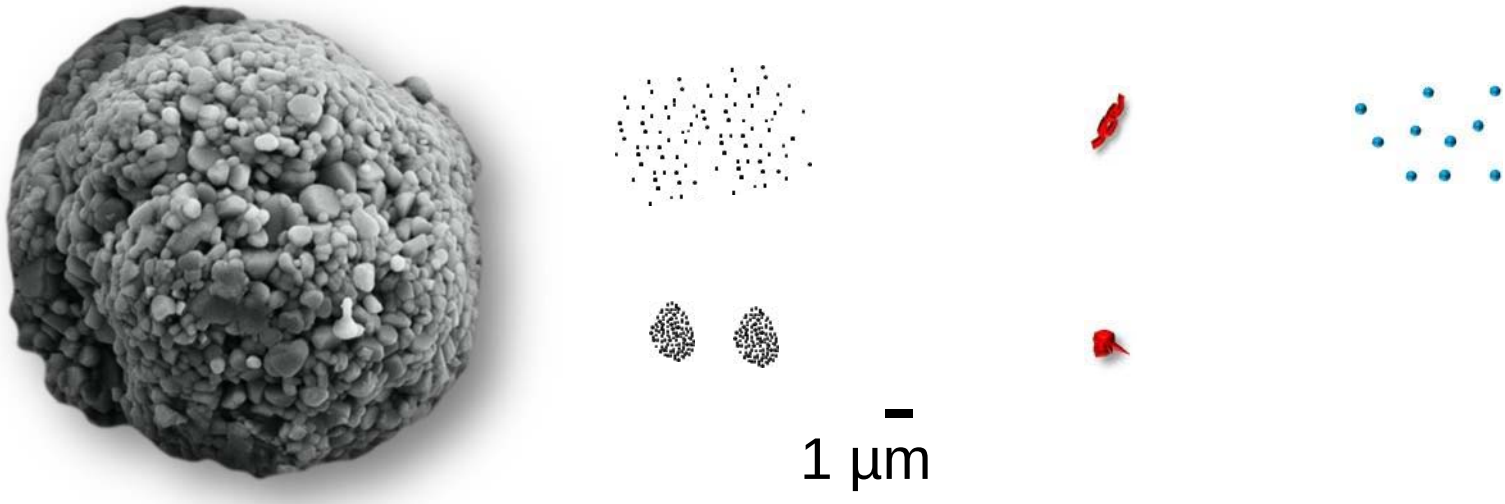
- Typical cathode paste formulation based on NMC

NMC (~ 10 µm)

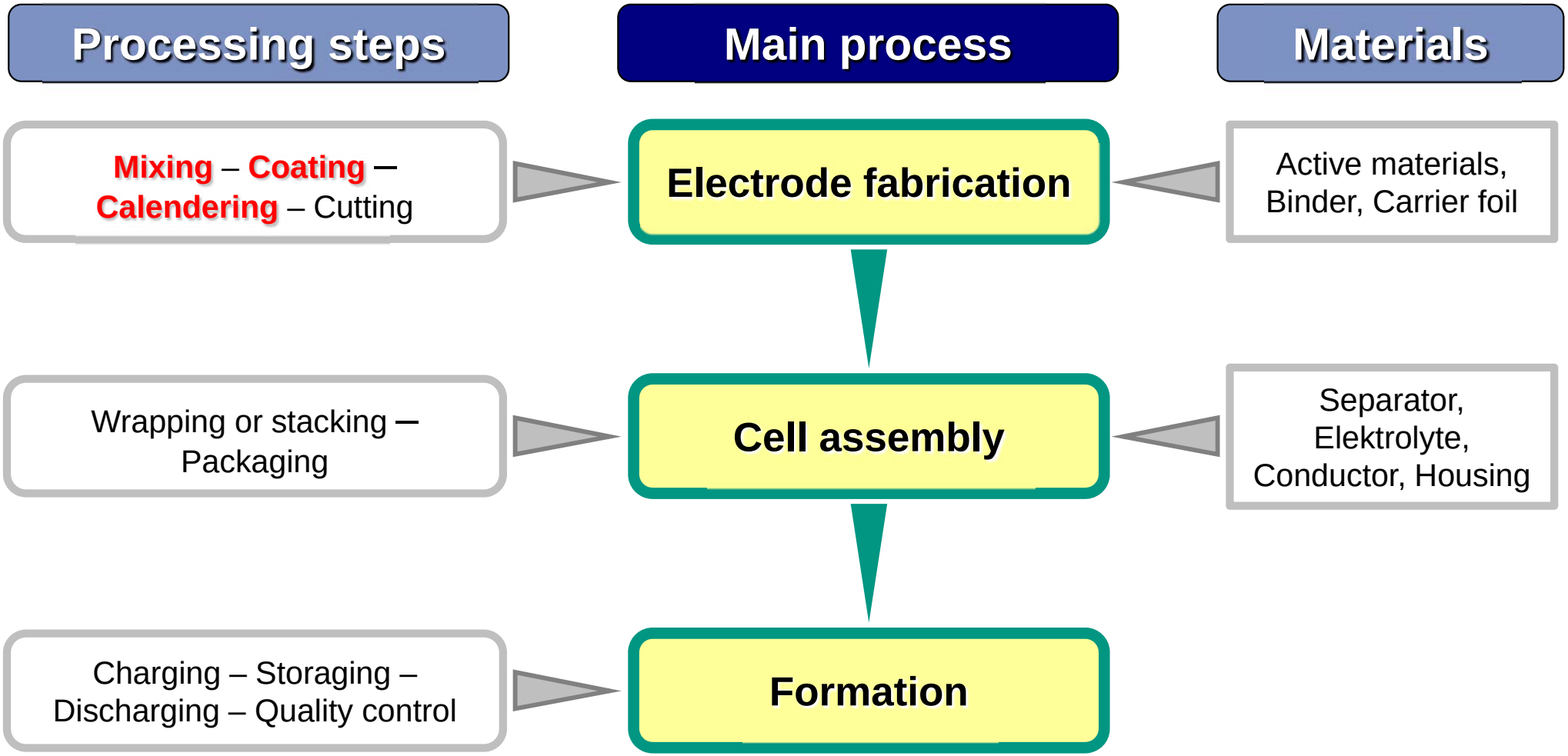
Carbon black (~ 40 nm)

CMC

SBR



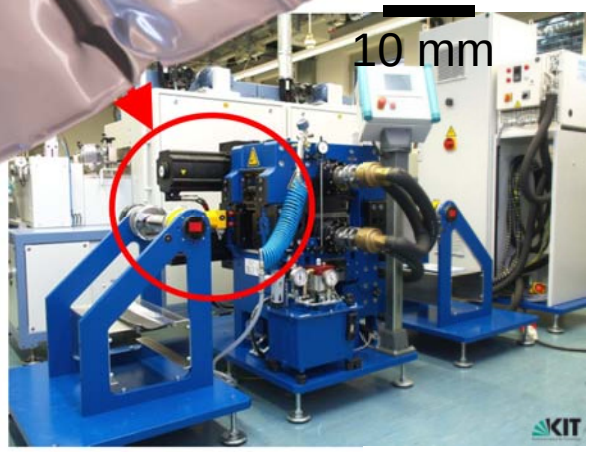
Cell manufacturing: from powder to cell



Cell manufacturing: from powder to cell

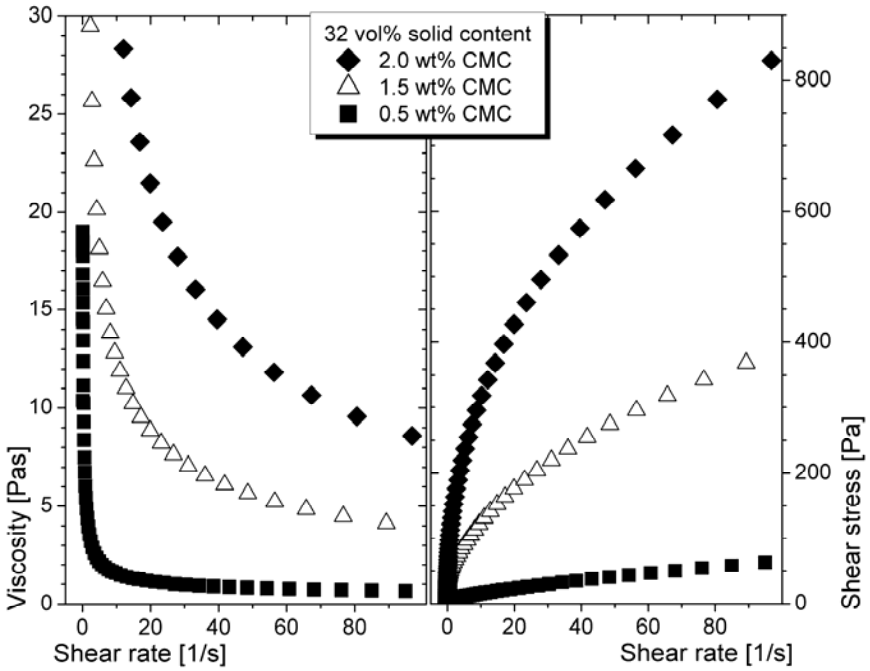
Mixing → Coating → Calendering → Cutting

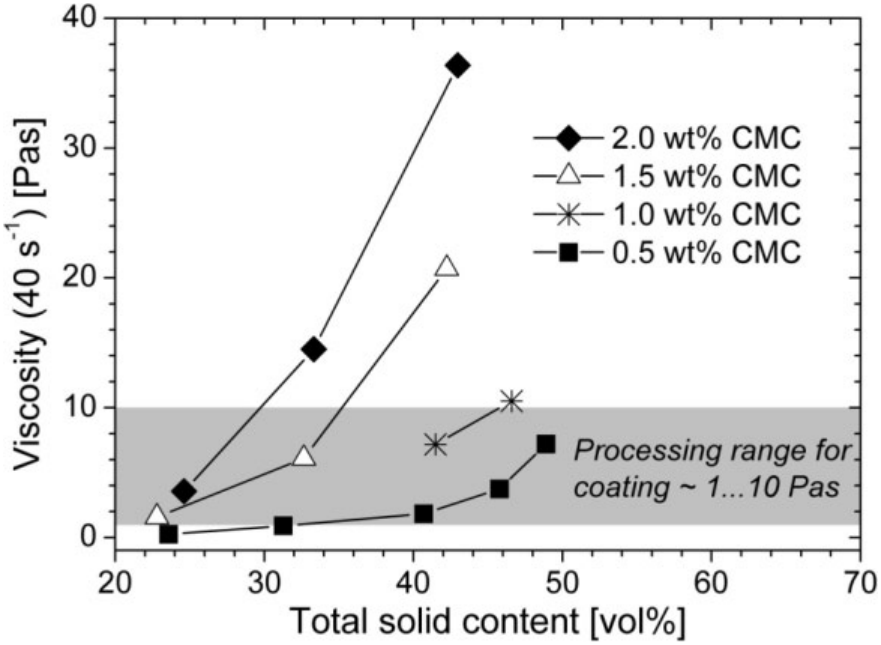
After stacking and packaging



Rheology of water-based NMC pastes

- Influence of **CMC** on paste rheology



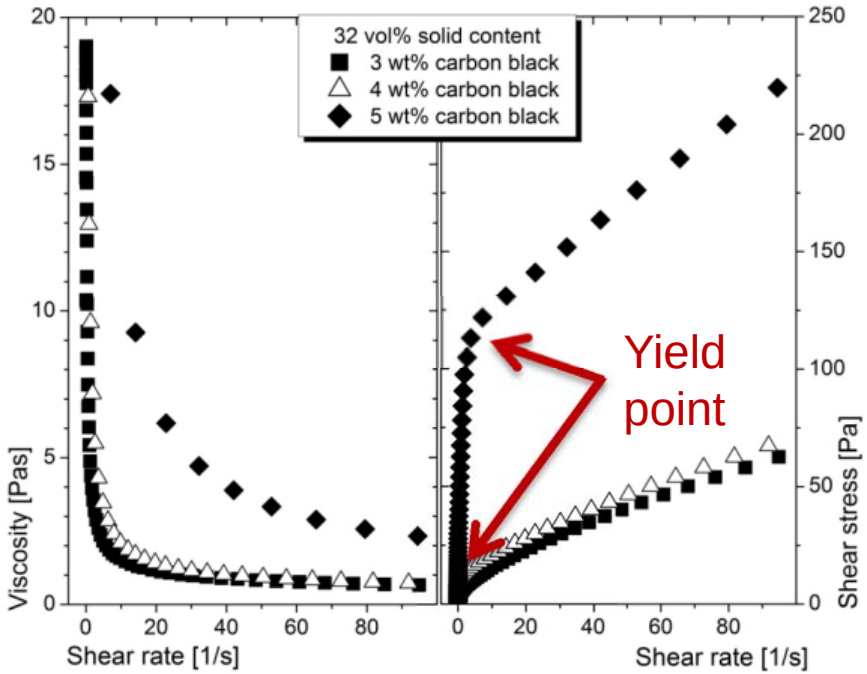


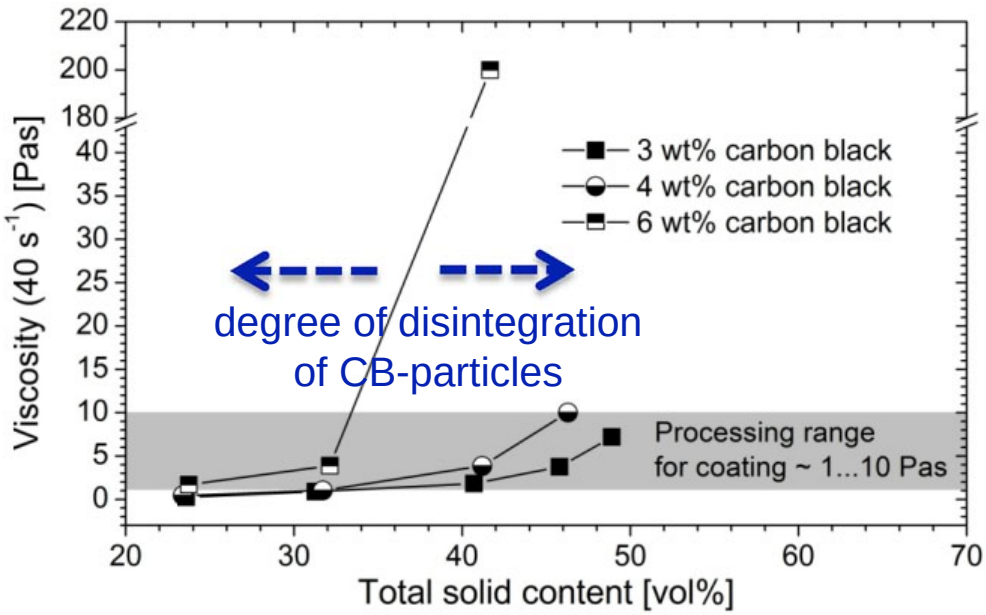
- Shear thinning flow behaviour
- Viscosity increase with increasing amount of CMC
- No distinctive yield point

- Thickening behaviour of CMC enables adjustment of paste viscosity
- Typical processing range for coating process: 1-10 Pas (at 40 s^{-1})

Rheology of water-based NMC pastes

- Influence of **carbon black (CB)** on paste rheology





- Distinctive yield point observed with addition of CB

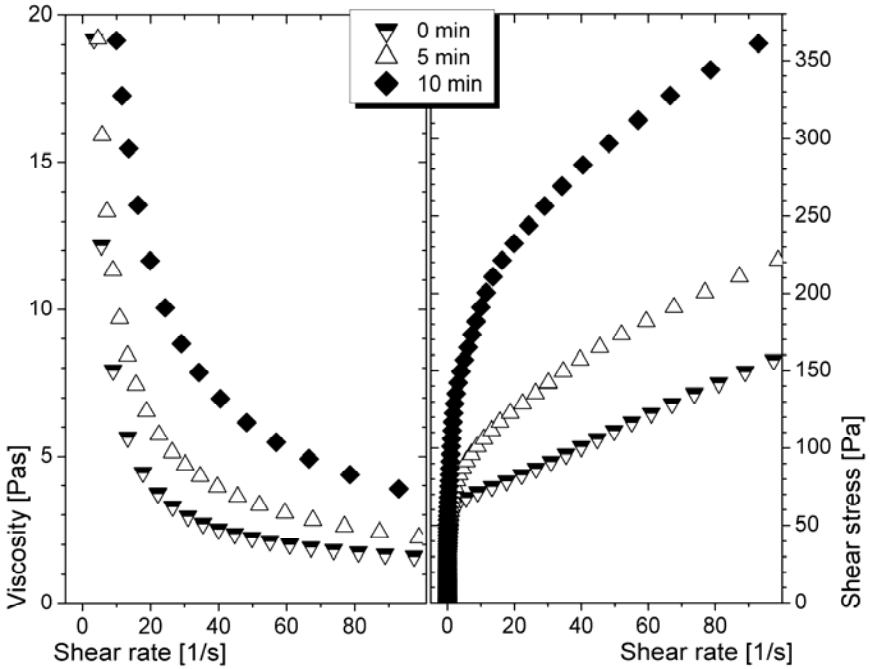
➔ Paste gelation at high CB amounts

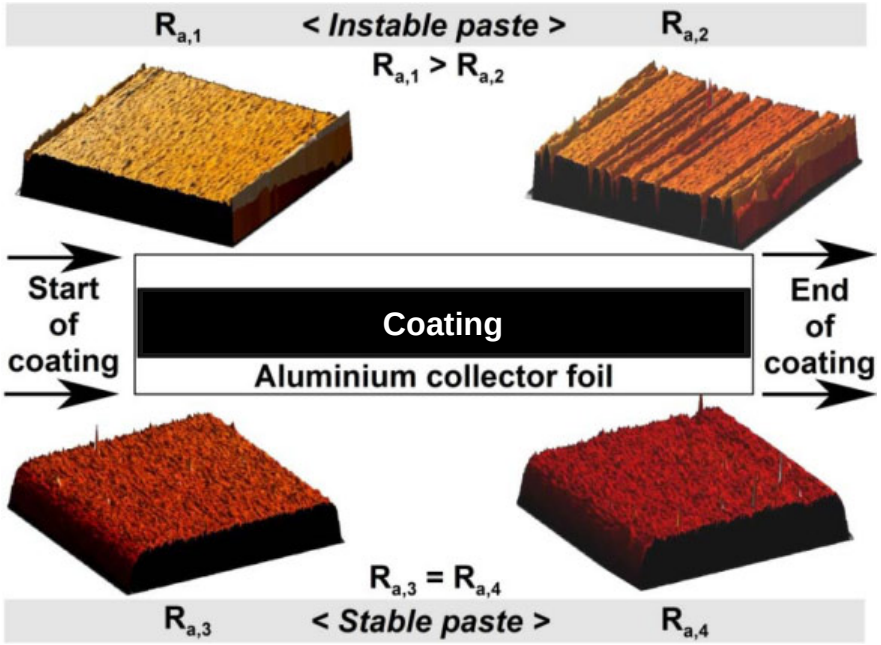
➔ Detrimental for coating quality

- Total solid content of pastes limited by amount of CB
- Threshold value depends on dispersing technique (disintegration degree of CB particles)

Rheology of water-based NMC pastes

- Paste stability and homogeneity

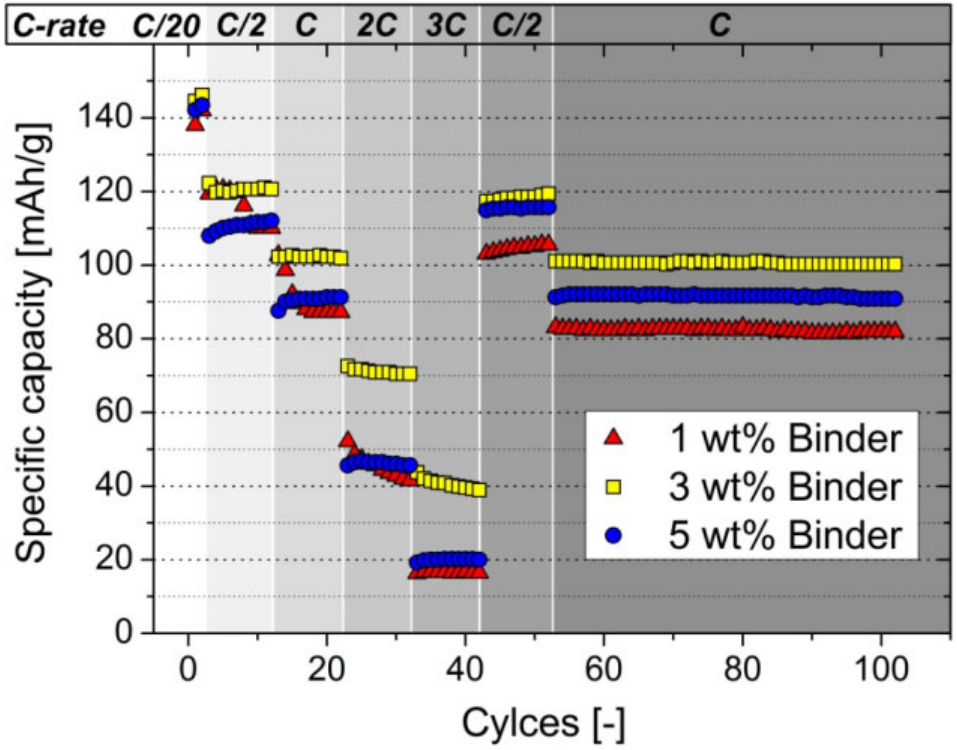


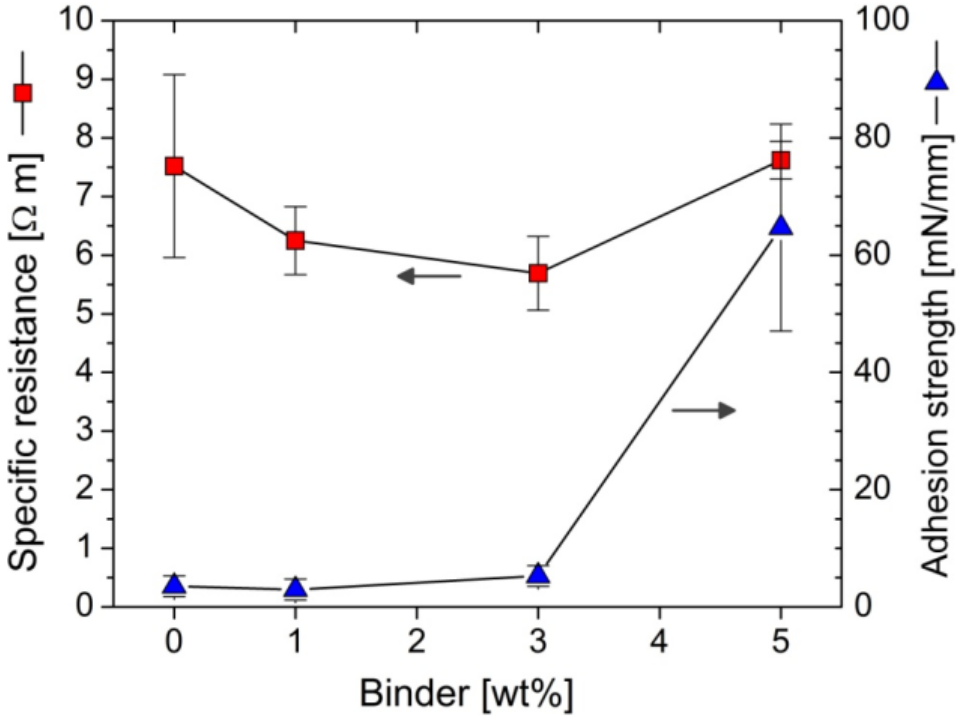


- Inappropriate combination of additives and process parameters may result in instable pastes
- Time-dependent flow behaviour
- Instable pastes ➔ inhomogeneous coatings (→ agglomeration)
- Surface roughness measurements allow evaluation of coating quality

Electrochemical properties of cells

CC-tests, 3.0-4.2 V, 20°C, charge/discharge at given C-rates

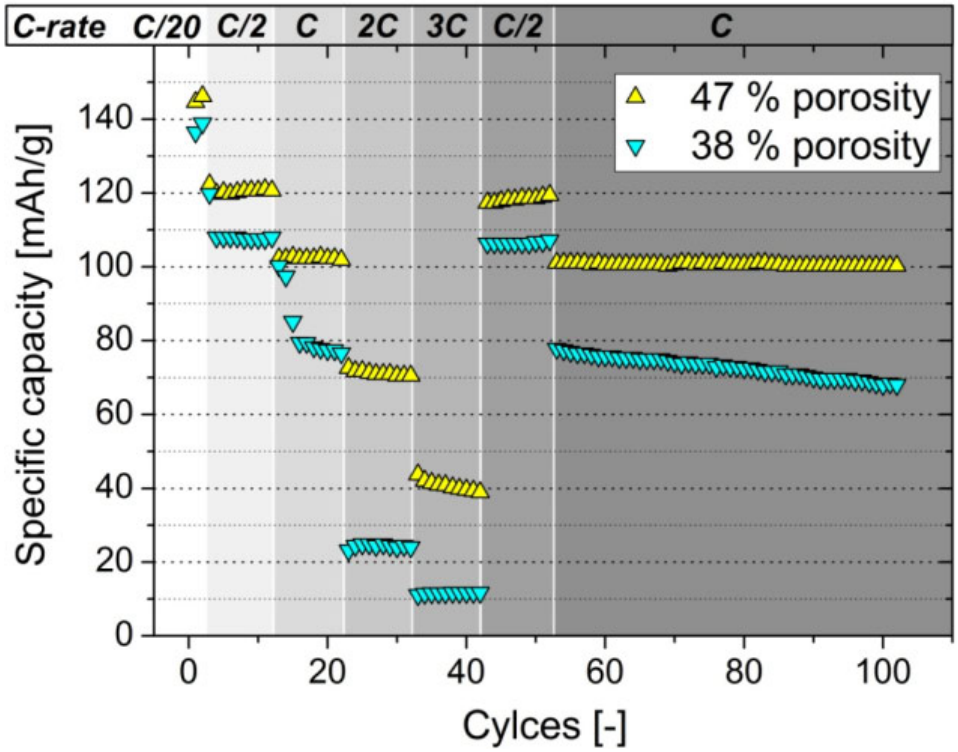




- Amount of latex binder influences cell performance, resistance and adhesion strength of coatings
- Addition of 3 wt% binder results in best cell performance and lowest specific resistance, but still poor adhesion ➔ optimization required (!)

Electrochemical properties of cells

CC-tests, 3.0-4.2 V, 20°C, charge/discharge at given C-rates



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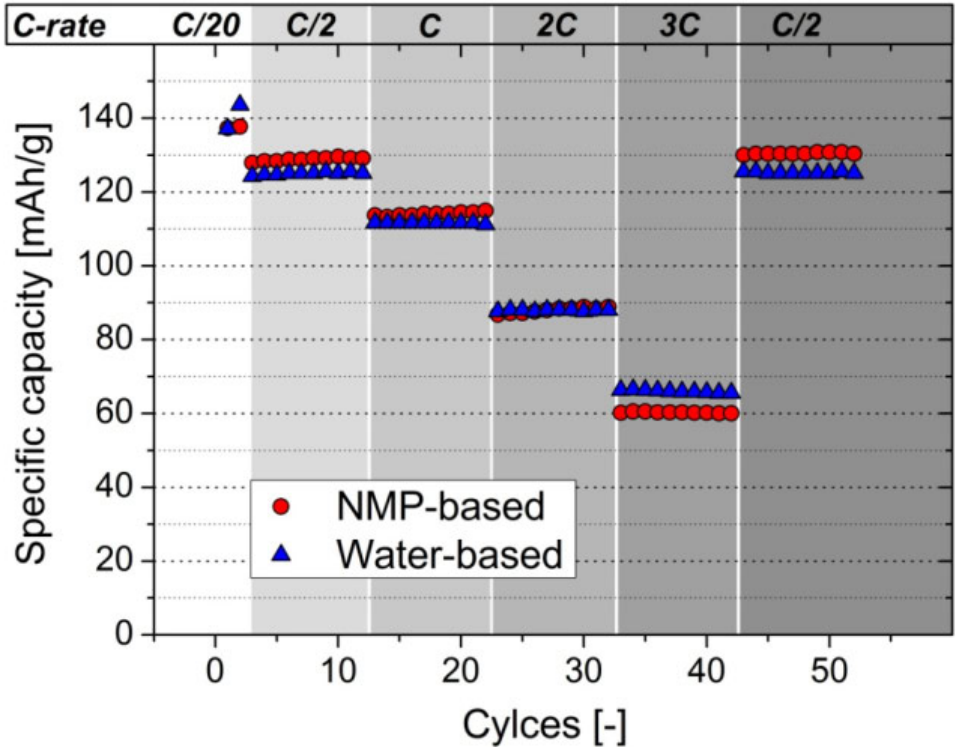


- Calendering conditions also affect cell performance (→ porosity)
- Water-based NMC cathodes exhibit cell performance comparable with NMP-based ones

Summary

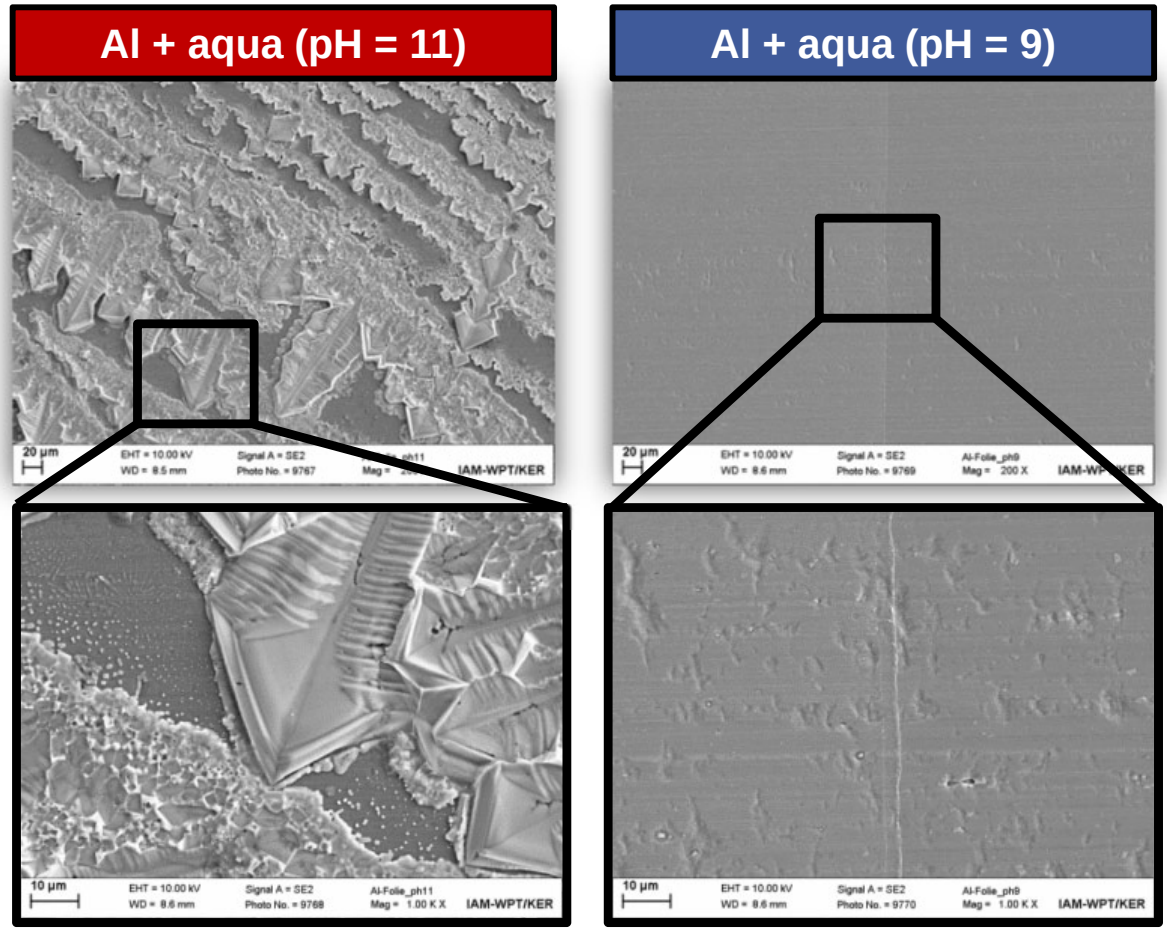
- Water-based NMC cathode pastes successfully prepared
- Paste stability and coating quality depends on type and amount of inactive additives (CMC, carbon black, binder)
- Amount of applied latex binder affects cell performance, coat resistance and adhesion strength of coating on Al-foil
- Variation of calendering conditions result in change of coat porosity, which also affects cell performance
- Electrochemical properties of water-based NMC cathodes comparable to conventional NMP-based ones

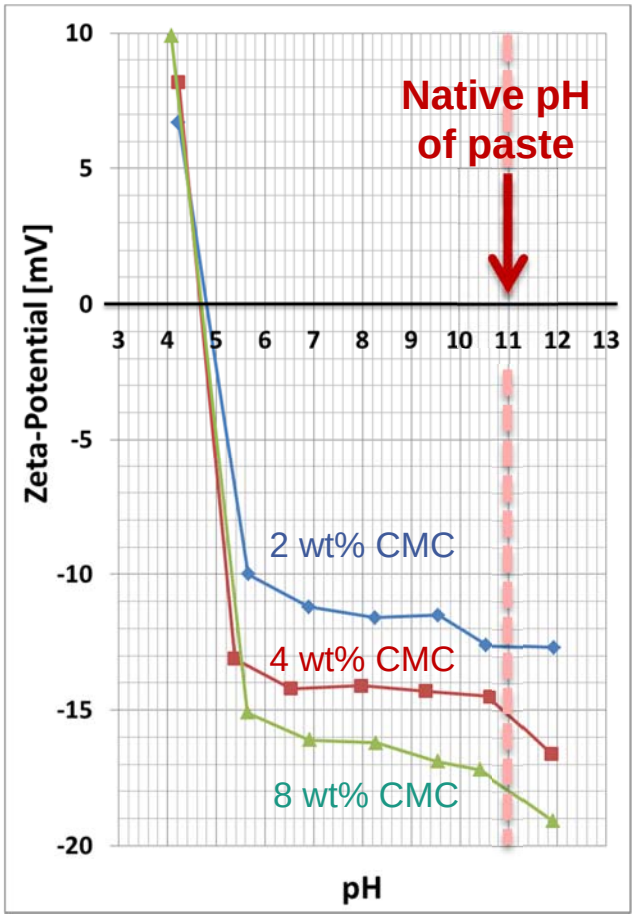
Outlook

- Amount / type of latex binder to be optimized in order to achieve good cell performance and high adhesion strength
- Calandering conditions to be optimized
- Influence of further types of additives (CMCs, binder,…) on paste homogeneity and cell performance to be studied
- Interaction of NMC with water to be studied
- Long-term cell performance (>> 100 cycles) to be tested
- Influence of paste pH value on corrosion of current collector foil (Al) to be considered

Outlook

- Influence of paste pH value on corrosion of current collector foil (Al) to be studied





Acknowledgements

- Helmholtz Association of German Research Centres for financial support
- C. Brösicke (IAM-WPT) for his support in cell assembling and electrochemical characterization

Thank you for your attention