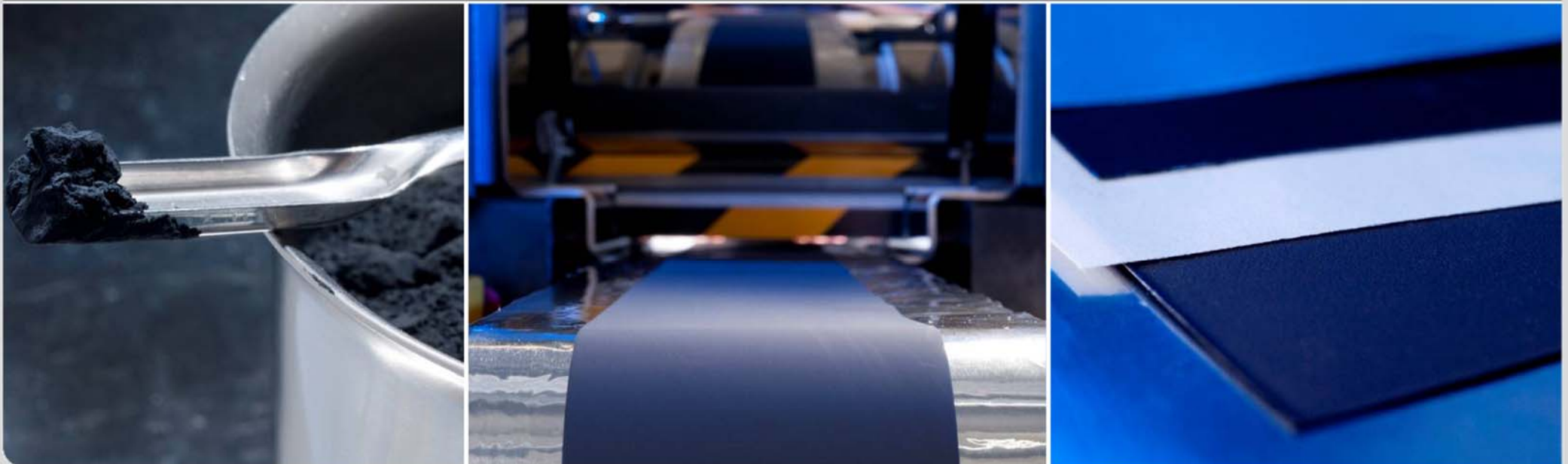


Processing of water-based electrode pastes for lithium nickel manganese cobalt oxide (NMC) batteries

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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



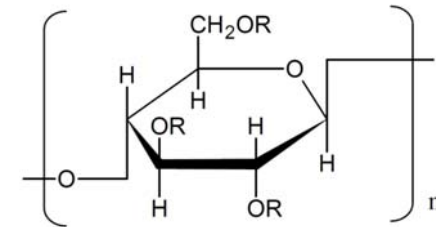
Issues of water-based processing

- 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)**

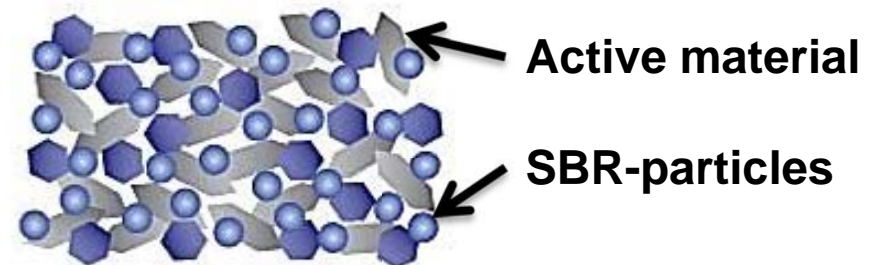
Thickening, dispersing and adhesive agent



- **Rubber or latex polymers**

e.g. Styrene-Butadiene-Rubber (SBR)

Binder, adhesive agent

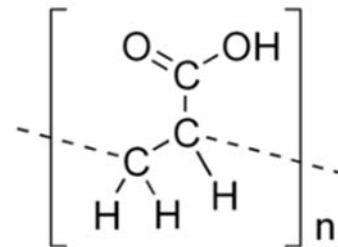


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- **Polyacrylic acid (PAA)**

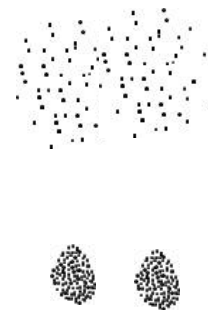
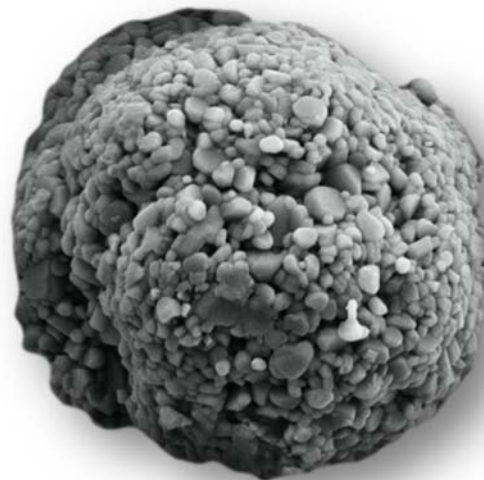
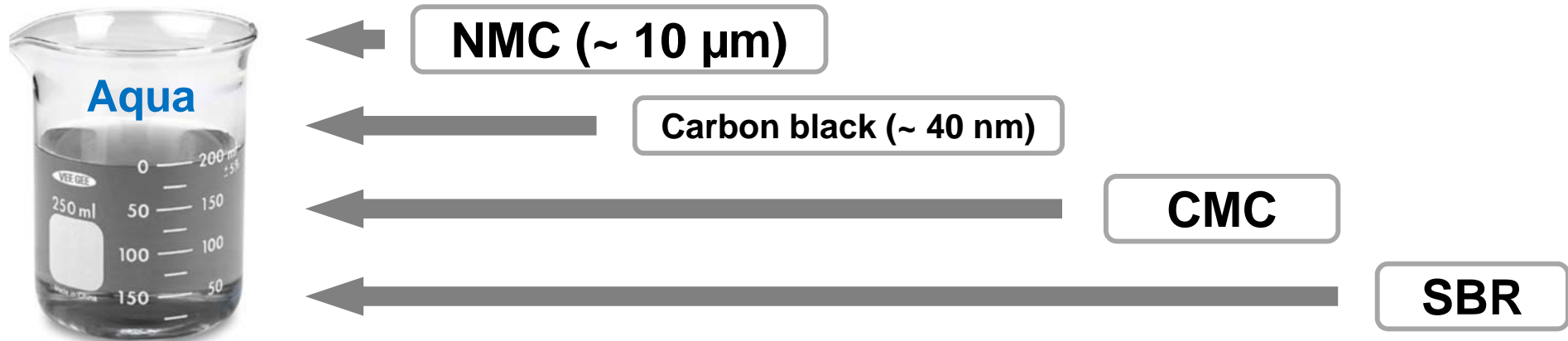
Dispersing agent

- ...



Preparation of water-based pastes

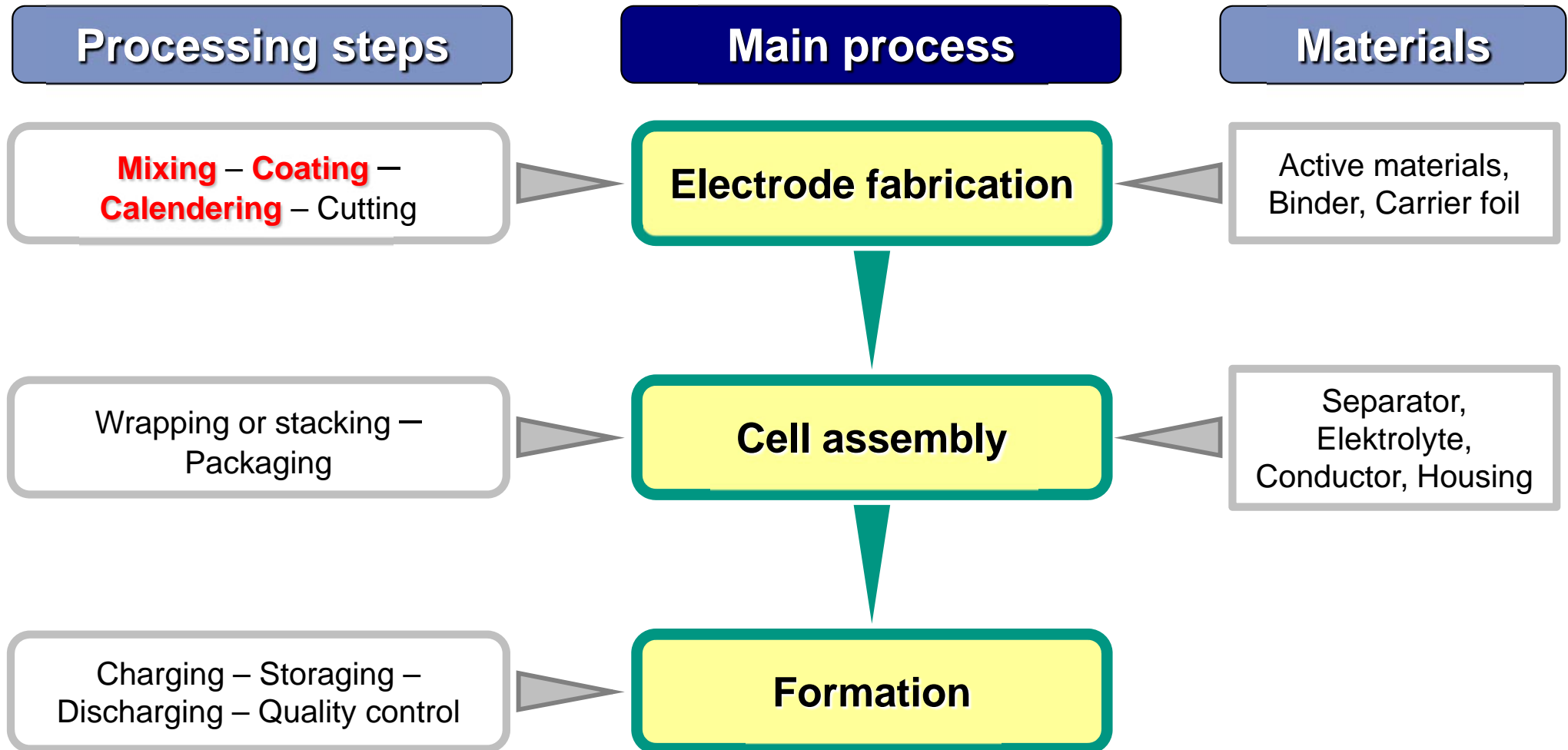
■ Typical cathode paste formulation based on NMC



1 μm



Cell manufacturing: from powder to cell



Cell manufacturing: from powder to cell

Mixing

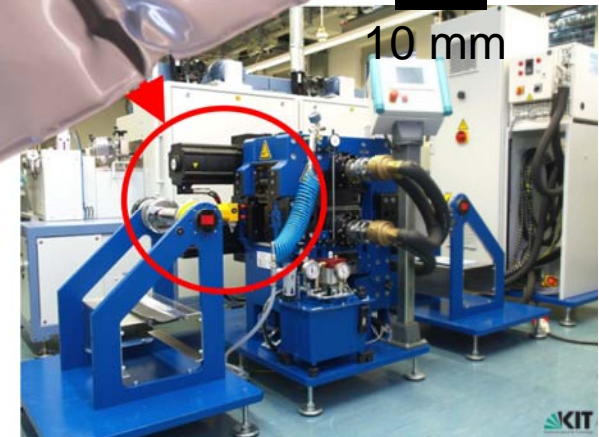
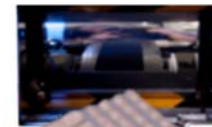
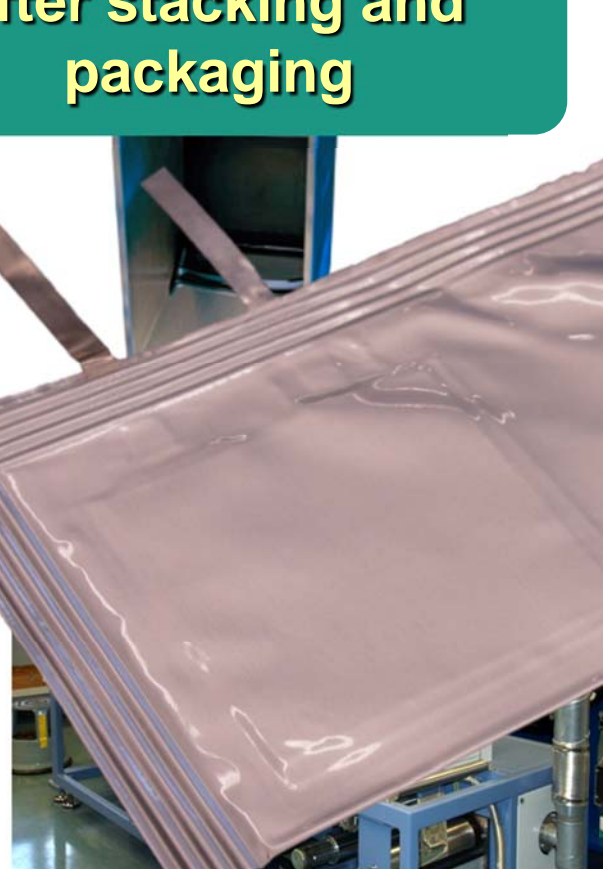
Coating

Calendering

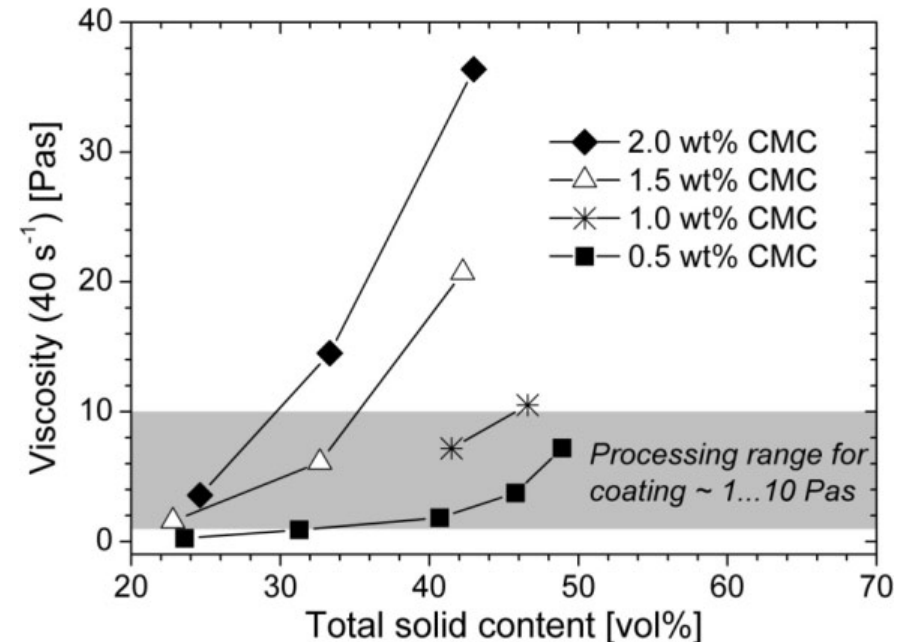
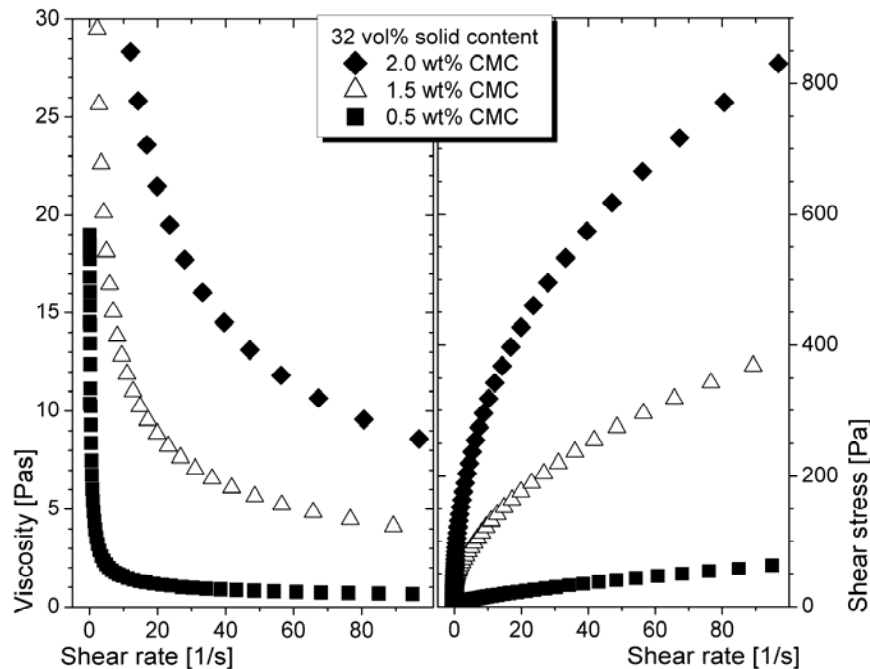
Cutting

After stacking and packaging

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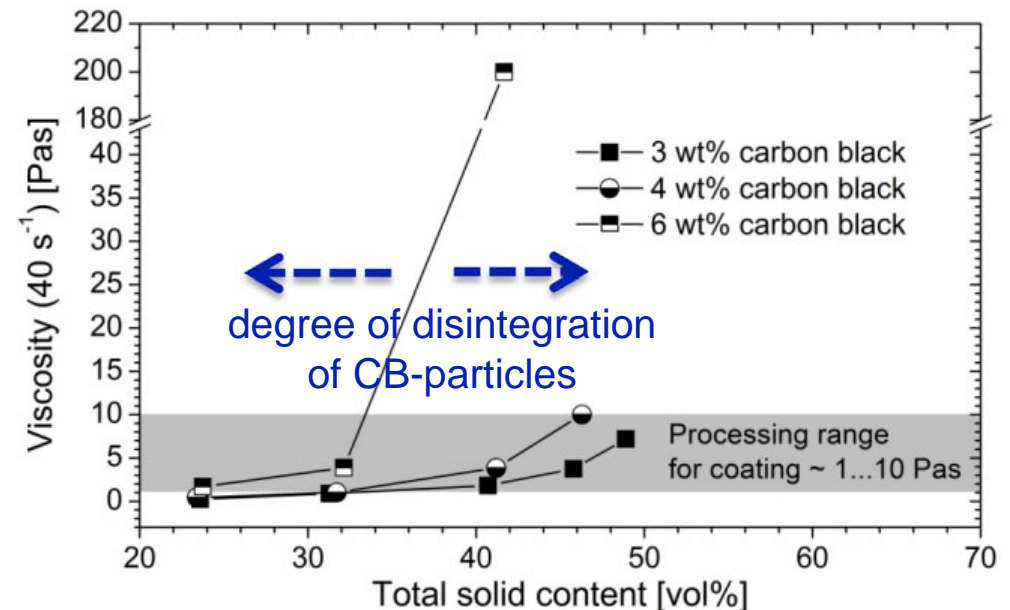
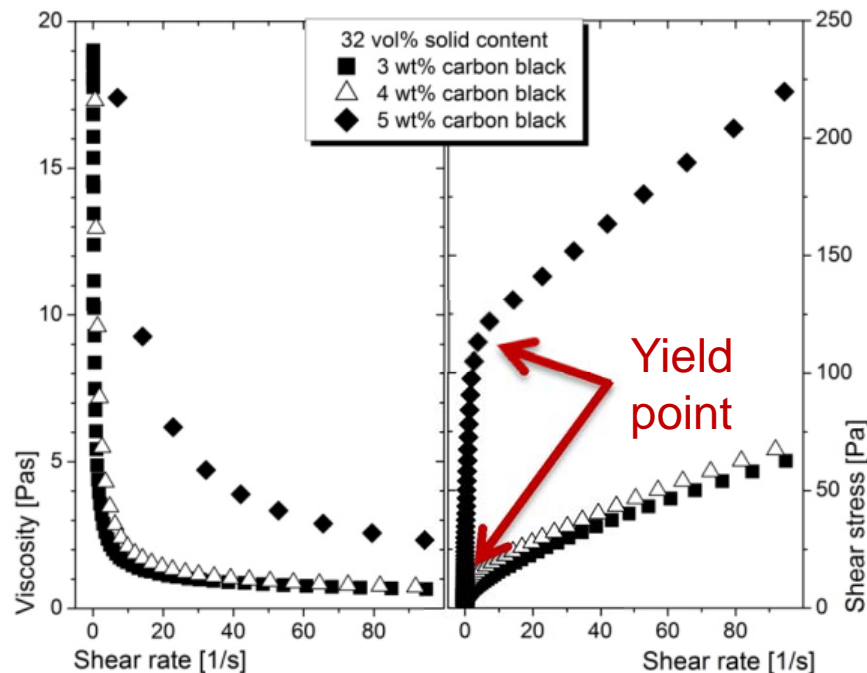
■ Influence of CMC on paste rheology



- 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

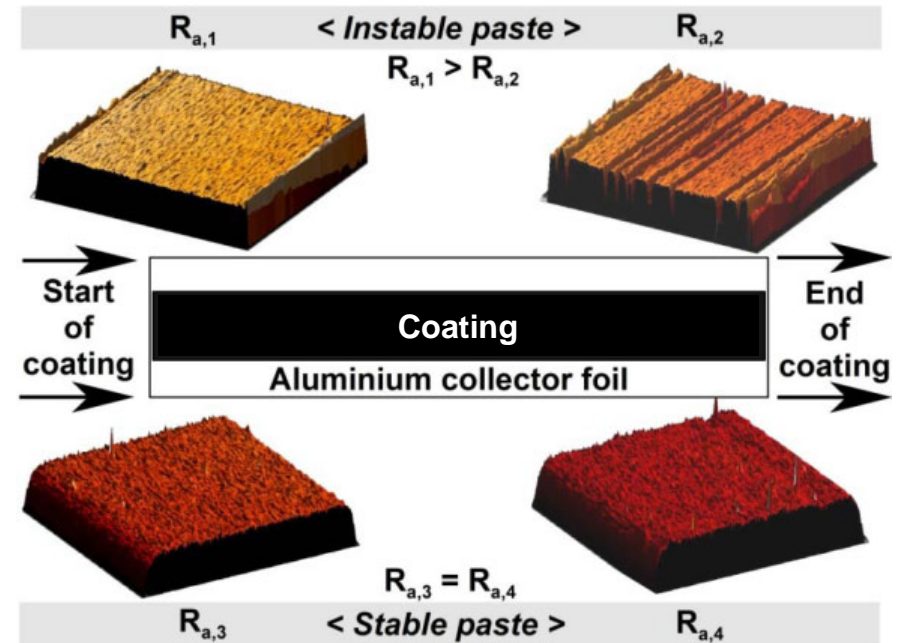
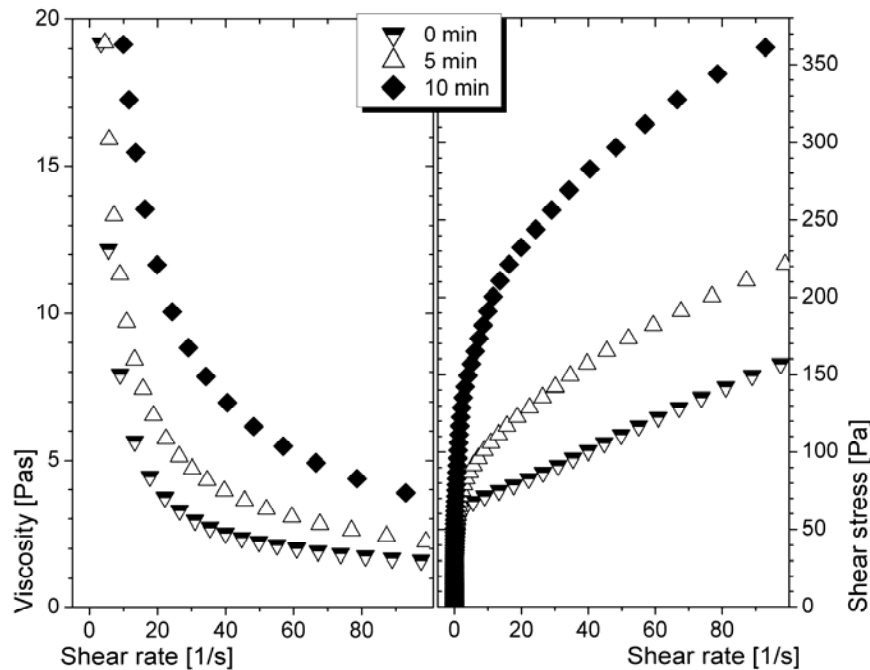
■ Influence of carbon black (CB) on paste rheology



- 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)

■ Paste stability and homogeneity

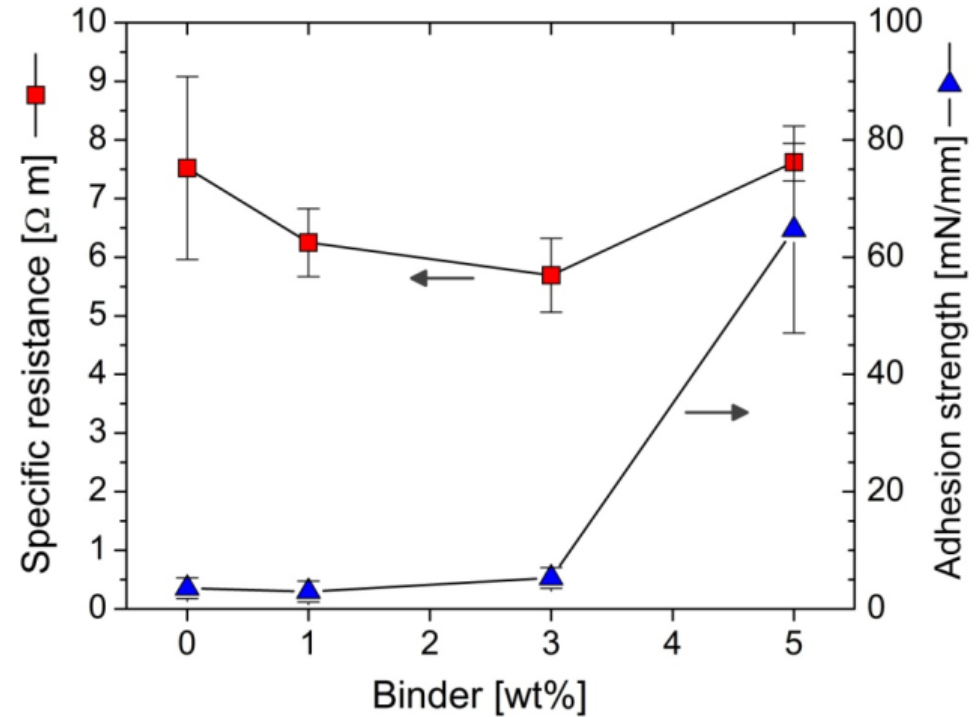
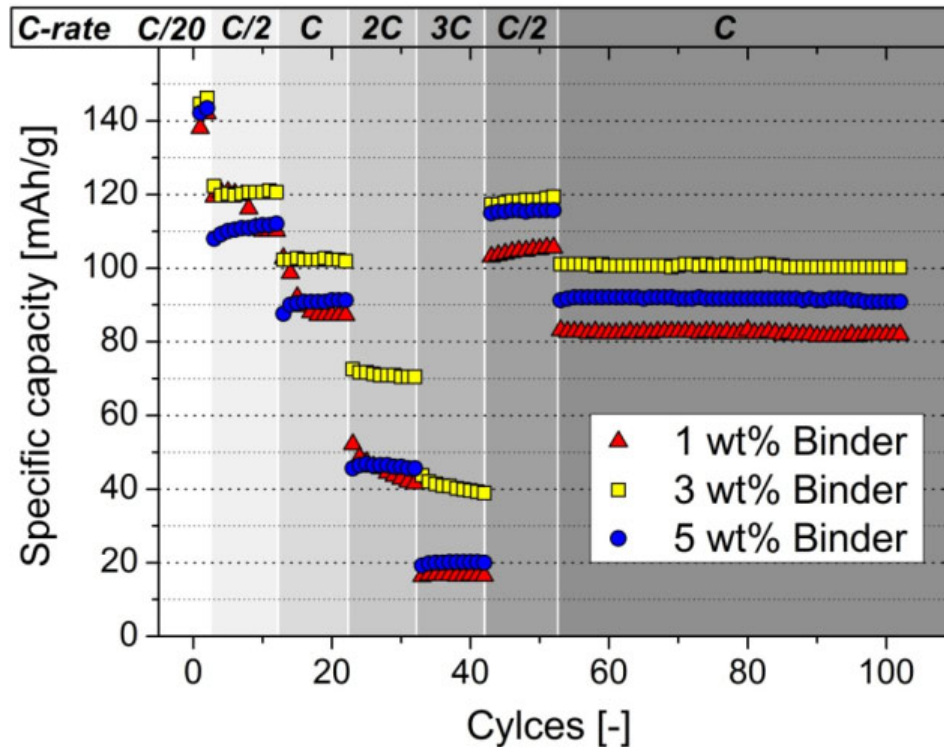


- Inappropriate combination of additives and process parameters may result in instable pastes
- Time-dependent flow behaviour

- Instable pastes → inhomogeneous coatings (→ agglomeration)
- Surface roughness measurements allow evaluation of coating quality

Electrochemical properties of cells

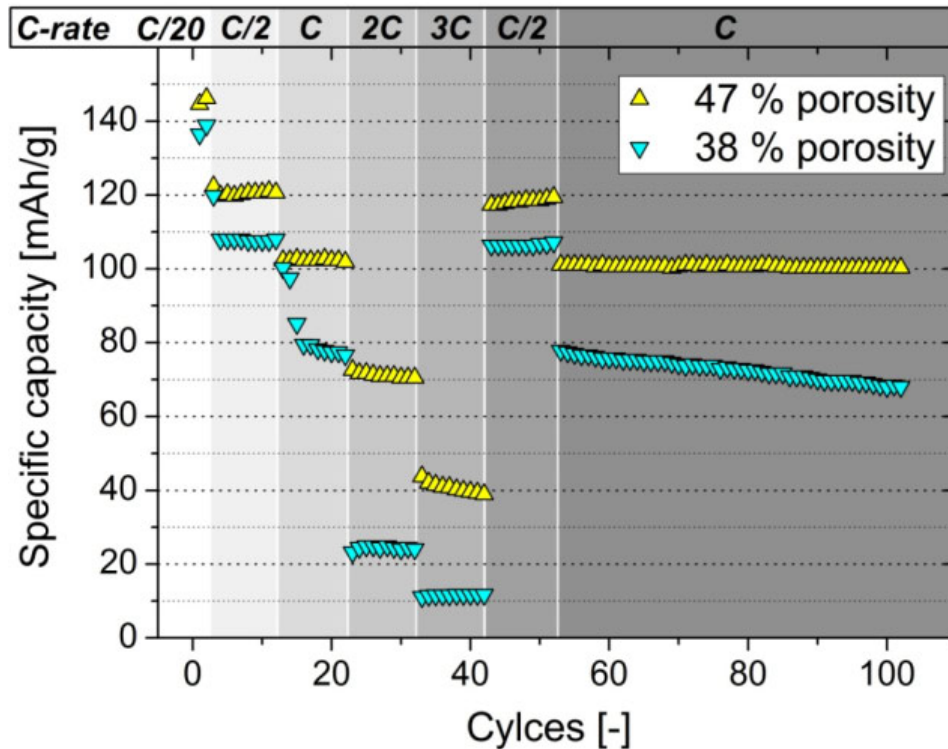
CC-tests, 3.0-4.2 V, 20°C, charge/discharge at given C-rates



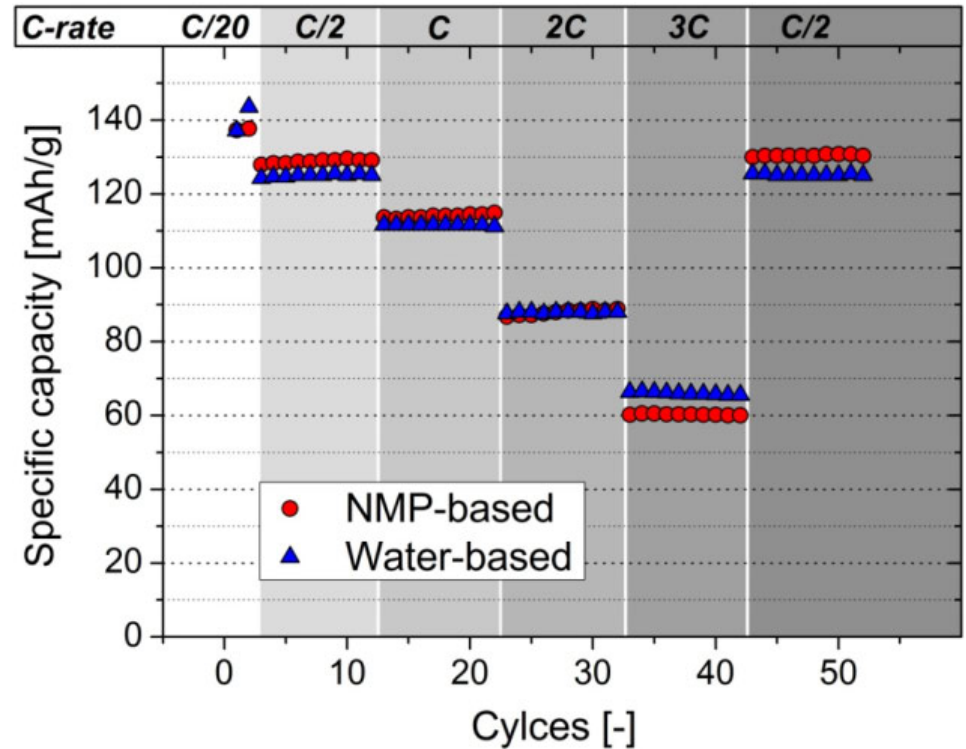
- Amount of latex binder influences cell performance, resistance and adhesion strength of coatings
- Addition of 3 wt% binder results in best cell performance and lowest specific resistance, but still poor adhesion → optimization required (!)

Electrochemical properties of cells

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



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

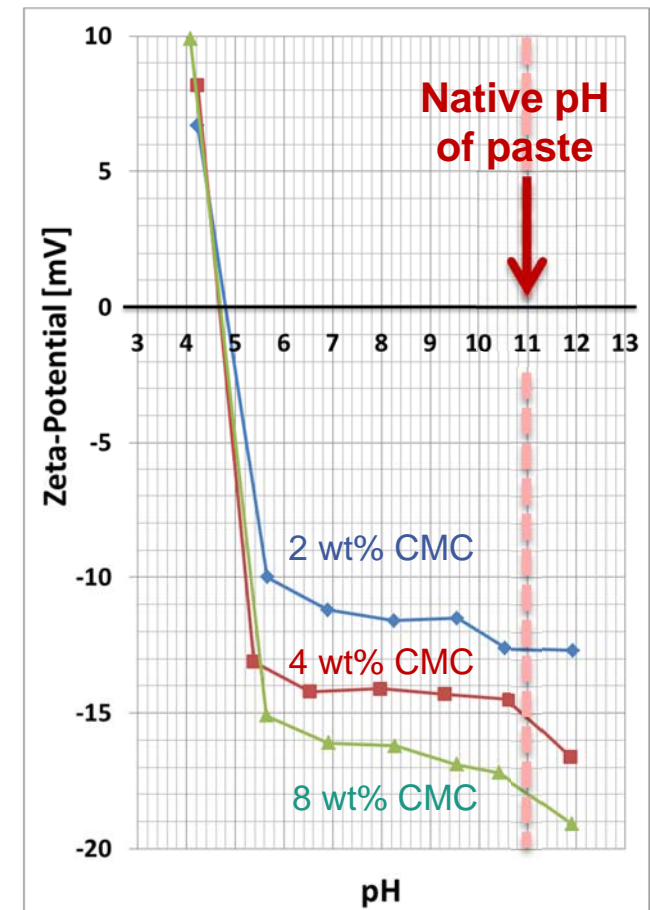
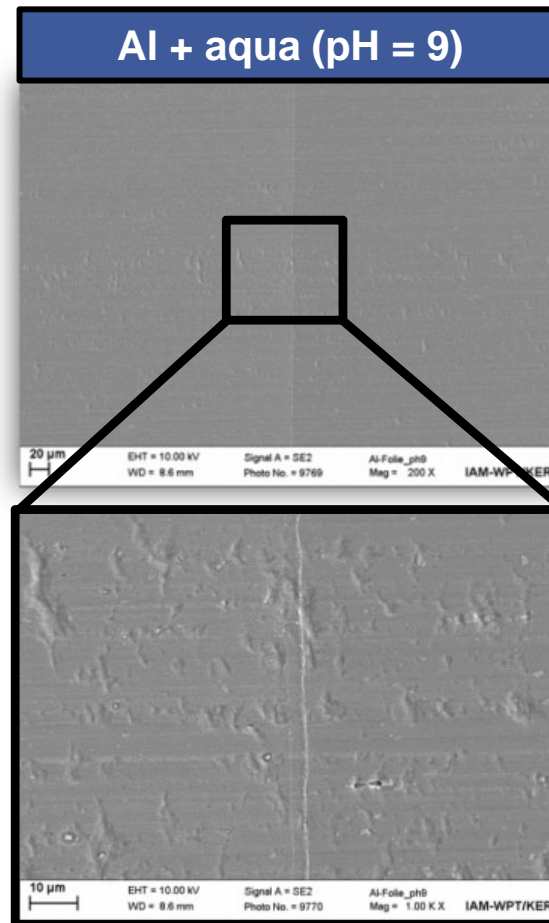
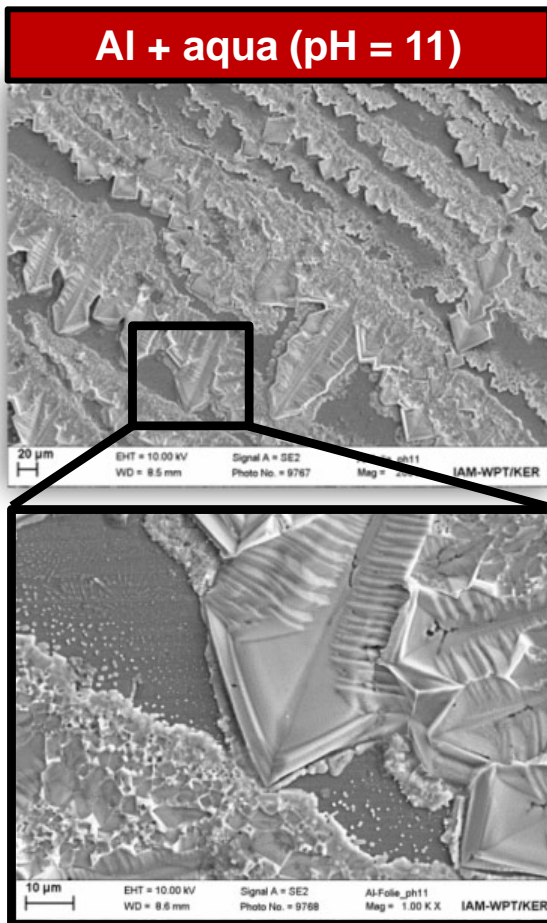


- Calendering conditions also affect cell performance (→ porosity)
- Water-based NMC cathodes exhibit cell performance comparable with NMP-based ones

- 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

- Amount / type of latex binder to be optimized in order to achieve good cell performance and high adhesion strength
- Calendering 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

- 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