

Lithium-sulphur batteries: Slot die coating and its comparison to comma bar coating

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Lithium-ion batteries are currently the leading technology in electric mobility and portable consumer electronics. However, this technology is already reaching its material specific limits in terms of achievable specific energy and requires scarce raw materials such as the expensive and ethically questionable cobalt. Concerning these aspects, the lithium-sulphur battery technology offers a reduction in material costs with additional higher specific energy. The active material sulphur, contained in the cathode, is not only economical due to low raw material prices but also electrochemically attractive. Sulphur has both, a high theoretical specific energy of approx. 2600 Wh/g and a high theoretical capacity of 1672 mAh/g. Due to high specific energies of lithium-sulphur battery, their use is particularly beneficial in gravimetrically critical applications, e.g. aircraft. Besides the material costs the production costs, especially for the electrode, must be reduced, for example by processing the electrode suspension with an extruder and by using a pre-metered slot die system for coating.

For continuous electrode production the water-based sulphur cathode suspension was produced with a twin-screw extruder followed by a degassing step. The particle size distribution was measured by laser diffraction and the rheological property of the suspension by shear rate test. The suspension was coated indirectly onto an aluminium foil using a comma bar or a slot die with a reverse roll coating system. The suspension coated on the aluminium foil was dried convectively. The local mass loading, adhesion strength, electrical conductivity as well electrochemical properties of the dried electrode were measured and related to the production parameters.

The diagram on the left side of figure 1 shows operating points of slot die coating modes as a function of the volume flow for a constant substrate speed and slot die gap as well as specific rheological prop-

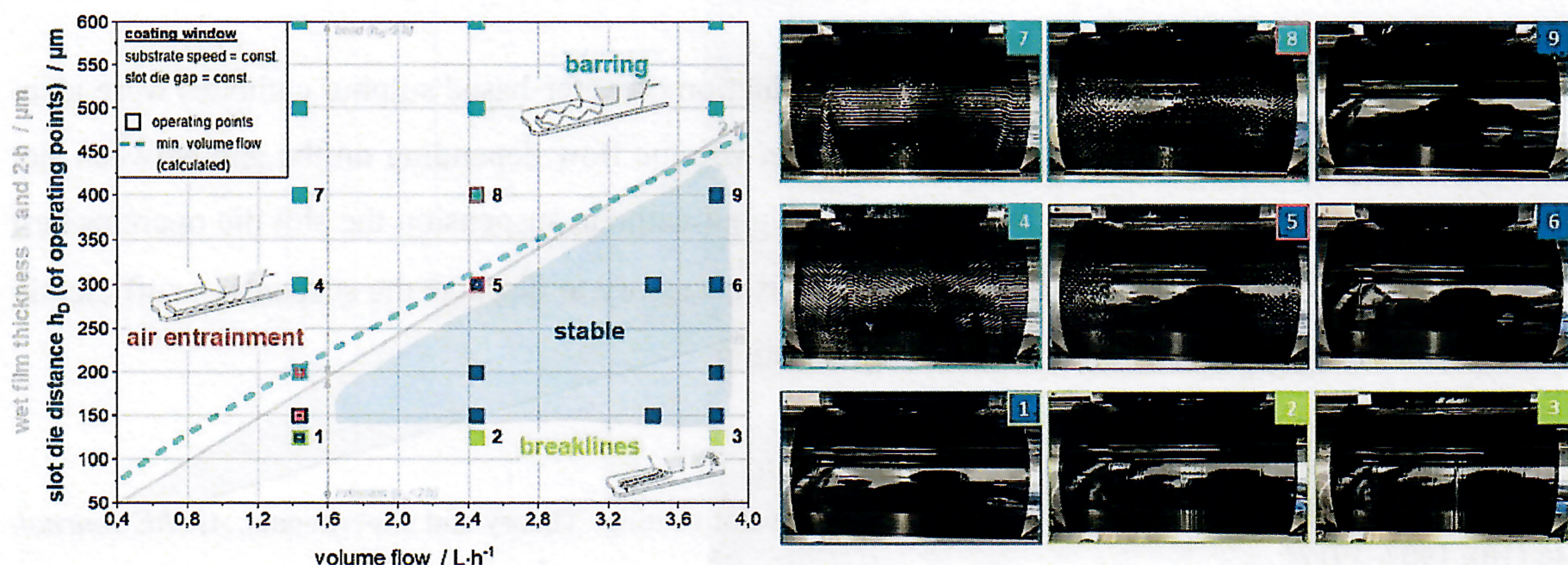


Figure 1: Coating window (left) with coating pictures of the displayed operating points (right; 1-9) (including illustrations from Tsuda (2010))

erties of the suspension. A stable coating window has been identified, while coating defects are displayed on the right side of the figure. In battery process technology a certain mass loading, which depends on the wet film thickness and therefore also on the volume flow, is to be coated on the electrode. Knowledge of the operating parameter setting of the slot die system is important to achieve defined mass loadings. Thus, the diagram also includes a function that specifies the minimum volume flow to obtain a stable coating for a given distance between slot die lips and roller. Equation (1) is based on the equation of low-flow limit (Carvalho and Kheshgi, 2000), which was modified according to the volume flow. It describes the stability of the downstream meniscus.

$$\dot{V}_{\min} = \frac{h_D \cdot B \cdot u_W}{\left(1,5 \cdot Ca^{-\frac{2}{3}}\right) + 1} \quad \text{with} \quad Ca = \frac{K \cdot u_W^n}{\sigma \cdot h_D^{n-1}} \quad (1)$$

h_D = gap between slot die and roller
 B = slot die width
 Ca = capillary number
 u_W = roll speed
 K = constant of power law model $\eta = K \cdot \dot{\gamma}^{n-1}$
 n = exponent of power law model
 η = viscosity
 σ = surface tension

As shown in Figure 1, this function is the boundary to the coating defect of barring. This defect occurs, if an operating point was selected with a large gap between slot die and roller, so that the gap cannot be bridged by suspension continuously. As expected, this phenomenon increases with decreasing volume flow. At low volume flows, this effect is superimposed by air entrainment, which always occurs first before a mixture of both effects or only barring occurs. This is caused by the upstream meniscus, through which air enters the slot die gap. If an operating point with a small gap between slot die and roller is selected, breaklines occur, which are caused by a higher volume flow and low rotational roller speed, whereby more mass reaches the gap without being able to be carried away directly. The result is suspension dripping out of the upstream meniscus and destabilization of the downstream meniscus.

In addition, the effects of a slight or intensive dispersion on the coating pattern were investigated. As expected, a slight dispersion resulted in a broad particle size distribution with large particles and agglomerates, which caused a poor coating performance due to uncoated stripes.

Further investigations regarding the comparison between comma bar and slot die showed that the comma bar has less sharp coating edges than the slot die and a poorer longitudinal distribution of mass loading. The comparison also shows no influence on electrochemical properties in coin cells.

Limitations of the slot die system regarding the production of water-based sulphur cathodes were identified and defined within a formula for the minimum volume flow depending on the gap between slot die and roller. With the processed shear thinning sulphur cathode suspension the slot die operates best in extrusion coating mode ($h_D < 2h$). Moreover, it is necessary to disperse the suspension sufficiently to prevent blockages and uncoated stripes on the coating.

References

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