Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Prospects on production technologies and manufacturing cost of oxide-based all-solid-state lithium batteries

Supplementary information

S1. Technology evaluation – technical suitability

Table S 1: Pairwise comparison for weighting of criteria for SSE layer fabrication, filled by experts in ceramics processing.

	Material requirements		**Product requirements**			**Production requirements**			
	Thermal stability of coating material	Thermal stability of substrate	Error rate	Layer density	Geometry	Environment	Throughput	Sum	**Normalized** (w_i)
Thermal stability of coating material		2	1	1	2	2	2	10	**3**
Thermal stability of substrate	0		0	0	2	1	2	5	**2**
Error rate	1	2		1	2	2	2	10	**3**
Layer density	1	2	1		2	2	2	10	**3**
Geometry	0	0	0	0		0	1	1	**1**
Environment	0	1	0	0	2		2	5	**2**
Throughput	0	0	0	0	1	0		1	**1**

Table S 2: Decision matrix for SSE layer fabrication, filled by experts in ceramics processing.

	Material requirements			**Product requirements**				**Production requirements**			**Score / ranking**
	Thermal stability of coating material	Thermal stability of substrate	M_{T_j}	Error rate	Layer density	Geometry	M_{T_j}	Environment	Throughput	M_{T_j}	
Weighting	3	2		3	3	1		2	1		
Vacuum slip casting	3	3	**3.00**	3	0	3	**1.71**	3	2	**2.67**	35
Tape casting	3	3	**3.00**	2	0	3	**1.29**	3	3	**3.00**	33
Screen Printing	3	3	**3.00**	2	0	3	**1.29**	3	3	**3.00**	33
Roller coating	3	3	**3.00**	2	0	3	**1.29**	3	2	**2.67**	32
Elektrophoretic deposition	3	3	**3.00**	2	0	3	**1.29**	3	1	**2.33**	31
Aerosol deposition methods	2	3	**2.40**	2	2	3	**2.14**	1	0	**0.67**	29
Wet powder spraying	3	3	**3.00**	1	0	3	**0.86**	3	1	**2.33**	28
Extrusion	3	3	**3.00**	0	0	3	**0.43**	3	3	**3.00**	27
VLPPS	0	2	**0.80**	3	3	3	**3.00**	0	1	**0.33**	26
Dip Coating	3	3	**3.00**	0	0	3	**0.43**	3	0	**2.00**	24
Spin Coating	3	3	**3.00**	0	0	3	**0.43**	3	0	**2.00**	24
LPPS	0	2	**0.80**	3	2	3	**2.57**	0	2	**0.67**	24
APS	1	3	**1.80**	2	0	3	**1.29**	2	2	**2.00**	24
CVD	0	2	**0.80**	3	3	0	**2.57**	0	0	**0.00**	22
EVD	0	0	**0.00**	3	3	3	**3.00**	0	0	**0.00**	21
High-velocity oxyfuel spraying	1	0	**0.60**	2	1	3	**1.71**	2	2	**2.00**	21
Flame spraying	1	3	**1.80**	0	0	3	**0.43**	2	2	**2.00**	18
Pulsed Laser Deposition	2	2	**2.00**	0	1	1	**0.57**	0	0	**0.00**	14
PVD-Electric beam	1	1	**1.00**	0	1	3	**0.86**	0	0	**0.00**	11
PVD-Sputtering	1	1	**1.00**	0	1	3	**0.86**	0	0	**0.00**	11

Table S 3: Pairwise comparison for weighting of criteria for cathode composite fabrication, filled by experts in ceramics processing.

	Material requirements		**Product requirements**			**Production require-ments**			
	Thermal stability of coating material	Thermal stability of substrate	Error rate	Layer den-sity	Geometry	Environ-ment	Throughput	Sum	**Normal-ized** (w_i)
Thermal stability of coating material		2	2	2	2	2	2	12	**3**
Thermal stability of substrate	0		1	2	2	1	1	7	**2**
Error rate	0	1		2	2	1	1	7	**2**
Layer den-sity	0	0	0		1	0	0	1	**1**
Geometry	0	0	0	1		0	0	1	**1**
Environ-ment	0	1	1	2	2		1	7	**2**
Throughput	0	1	1	2	2	1		7	**2**

Table S 4: Decision matrix for cathode composite fabrication, filled by experts in ceramics processing.

	Material requirements			Product requirements				Production requirements			Score / ranking
	Thermal stability of coating material	Thermal stability of substrate	M_{T_j}	Error rate	Layer density	Geometry	M_{T_j}	Environment	Throughput	M_{T_j}	
Weighting	3	2		2	1	1		2	2		
Tape casting	3	3	**3.00**	3	0	3	**2.25**	3	3	**3.00**	36
Screen printing	3	3	**3.00**	2	0	3	**1.75**	3	3	**3.00**	34
Extrusion	3	3	**3.00**	1	0	3	**1.25**	3	3	**3.00**	32
Roller coating	3	3	**3.00**	2	0	3	**1.75**	3	2	**2.50**	32
Vacuum slip casting	3	3	**3.00**	2	0	3	**1.75**	3	2	**2.50**	32
Elektrophoretic deposition	3	3	**3.00**	2	0	3	**1.75**	3	1	**2.00**	30
Wet powder spraying	3	3	**3.00**	2	0	3	**1.75**	3	1	**2.00**	30
Atmospheric plasma spraying	1	3	**1.80**	3	2	3	**2.75**	2	2	**2.00**	28
Dip /Spin coating	3	3	**3.00**	0	0	3	**0.75**	3	0	**1.50**	24
Aerosol deposition methods	2	3	**2.40**	2	2	3	**2.25**	1	0	**0.50**	23
Flame spraying	1	3	**1.80**	1	1	3	**1.50**	2	2	**2.00**	23
High-velocity cxy-fuel spraying	0	1	**0.40**	3	2	3	**2.75**	2	2	**2.00**	21
LPPS	0	2	**0.80**	3	3	3	**3.00**	0	2	**1.00**	20
VLPPS	0	2	**0.80**	3	3	3	**3.00**	0	1	**0.50**	18
EVD	2	2	**2.00**	0	3	3	**1.50**	0	0	**0.00**	16
CVD	2	2	**2.00**	0	3	0	**0.75**	0	0	**0.00**	13
Pulsed Laser Deposition	2	2	**2.00**	0	1	1	**0.50**	0	0	**0.00**	12
PVD-Electric beam	1	1	**1.00**	0	1	3	**1.00**	0	0	**0.00**	9
PVD-Sputtering	1	1	**1.00**	0	1	3	**1.00**	0	0	**0.00**	9

S2. Technology evaluation – technology maturity

Questionnaire to determine the maturity of aerosol coating for the production of ceramic solid state cells

General information

Technology designation	Responsible person, function, organization/institute, department	Date
T_2 Aerosol Coating		

General Data

Producible layer thicknesses ________________

Temperature input on substrate ________________

Temperature input on coating material ________________

The following compression/heat treatment step is necessary ________________

1. fundamental research

Maturity level description

This level is the lowest on the scale of maturity. It contains the basic mechanisms and principles of action as well as mathematical and physical descriptions and sketches. This results in theoretical concepts.

1.1

Is there a technical description of the (functional) principle of aerosol coating in the form of patents, publications, dissertations, with mathematical formulas, physical interrelations, geometry or material data, etc.?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

1.2

Are the following process parameters known? Starting material, carrier gas and its consumption, nozzle settings, gas system, vibration system?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

1.3

Is there a technical production application area for the aerosol coating?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

1.4

Are there several different applications (in several industries) for aerosol coating? If so, which ones?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

1.5

Is the future use of the technology in an industrial environment still speculative?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

1.6

Notes:

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

Figure S 1: Exemplary questionnaire to determine technology maturity – stage 1 (adapted and translated from Schindler (2015)[1])

Questionnaire to determine the maturity of aerosol coating for the production of ceramic solid state cells

General information

Technology designation	Responsible person, function, organization/institute, department	Date
T2 Aerosol Coating		

2. Feasibility study

Maturity level description

In this stage, the theories from stage 1 are tested using simple experiments or simulations. The aim is to identify influencing factors and requirements.

2.1

Are the essential functions of the system components (vacuum chamber system, gas system, vibration system, nozzle) known for the realization of the aerosol coating? Is it known what needs to be investigated experimentally or simulatively in order to demonstrate the feasibility of producing a cell layer?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

2.2

Are the essential conditions for a realization of the application identified?
(e.g. boundary conditions such as laws, environmental conditions, etc.).

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

2.3

Have the essential process-related requirements for the implementation of the application been identified?
(e.g. starting material)

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

2.4

Have other technologies and resources been identified that are necessary to apply aerosol coatings?
(Possible resources: gas connection, chamber)

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

2.5

Has the technological feasibility of the individual function/subsystems been validated by fundamental experiments or analytically, e.g. by simulation?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

2.6

Notes:

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

Figure S 2: Exemplary questionnaire to determine technology maturity – stage 2 (adapted and translated from Schindler (2015)[1])

Questionnaire to determine the maturity of aerosol coating for the production of ceramic solid state cells

General information

Technology designation	Responsible person, function, organization/institute, department	Date
T2 Aerosol Coating		

3. Development of technology

Maturity level description

In this stage, the requirements and findings from stage 2 are checked. Alternative concepts are developed and the best concept selected. The result of the stage contains the essential information for the construction of the prototype.

No.	Question	Answer	Degree of fulfilment / uncertainty:
3.1	Were the influencing factors and requirements from level 2 documented and checked, including mathematical models, calculations or simulations?	☐ yes ☐ no ☐ uncertain	0 10 20 30 40 50 60 70 80 90 100
3.2	Are the essential parameters of the aerosol coating described? Is it known how to set the parameters for the individual functions? Parameters: Starting material, carrier gas and its consumption, nozzle settings, gas system settings and vibration system settings	☐ yes ☐ no ☐ uncertain	0 10 20 30 40 50 60 70 80 90 100
3.3	Are the essential interdependencies and interactions as well as possibilities of influencing the parameters (partly theoretically) known?	☐ yes ☐ no ☐ uncertain	0 10 20 30 40 50 60 70 80 90 100
3.4	Was the coarse process window of the technology formulated on the basis of calculations or simulations?	☐ yes ☐ no ☐ uncertain	0 10 20 30 40 50 60 70 80 90 100
3.5	Are the essential components and elements required to build a prototype/system known?	☐ yes ☐ no ☐ uncertain	0 10 20 30 40 50 60 70 80 90 100
3.6	Notes:	☐ yes ☐ no ☐ uncertain	0 10 20 30 40 50 60 70 80 90 100

Figure S 3: Exemplary questionnaire to determine technology maturity – stage 3 (adapted and translated from Schindler (2015)[1])

Questionnaire to determine the maturity of aerosol coating for the production of ceramic solid state cells

General information

Technology designation	Responsible person, function, organization/institute, department	Date
T_2 Aerosol Coating		

4. Technology demonstrator

Maturity level description

In stage 4, a prototype is built for the first time. This should validate the previously determined process parameters and functions of the technology. The functional testing takes place in a laboratory environment whose framework conditions are similar to the real environment.

	Question	Assessment
4.1	Was a functional prototype of the aerosol spraying system set up to describe the overall system?	yes ☐ no ☐ uncertain ☐ Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100
4.2	Have the main previously determined process parameters been validated?	yes ☐ no ☐ uncertain ☐ Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100
4.3	Have the main functions (e.g. supply of powder particles, homogeneous distribution of particles and carrier gas, transfer of particles through nozzle) of the system been validated?	yes ☐ no ☐ uncertain ☐ Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100
4.4	Was the prototype built in a sufficient environment that reflects the conditions of the real production environment?	yes ☐ no ☐ uncertain ☐ Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100
4.5	Is the applicability of the aerosol coating for the production of cell layers in a sufficiently similar environment proven?	yes ☐ no ☐ uncertain ☐ Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100
4.6	Notes:	yes ☐ no ☐ uncertain ☐ Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

Figure S 4: Exemplary questionnaire to determine technology maturity – stage 4 (adapted and translated from Schindler (2015)[1])

Questionnaire to determine the maturity of aerosol coating for the production of ceramic solid state cells

General information

Technology designation	Responsible person, function, organization/institute, department	Date
T2 Aerosol Coating		

5. Integration into equipment

Maturity level description

This stage involves the integration of the technology into an equipment in a laboratory environment under realistic conditions. The functions of the entire system and the process window can be tested and validated.

No.	Question	yes	no	uncertain	Degree of fulfilment / uncertainty (0–100)
5.1	Has the aerosol coating been implemented in an equipment in a laboratory environment under realistic conditions with set process parameters?	☐	☐	☐	0 10 20 30 40 50 60 70 80 90 100
5.2	Are the relevant interactions between the parameters in the prototype known?	☐	☐	☐	0 10 20 30 40 50 60 70 80 90 100
5.3	Are the interactions between the aerosol coating and upstream and downstream technologies (drying, sintering, cutting, etc.) within the production line known/recorded?	☐	☐	☐	0 10 20 30 40 50 60 70 80 90 100
5.4	Is a smooth production process possible when using aerosol coating in consideration of the given technology chain for the production of cell layers? If not, which production step is problematic?	☐	☐	☐	0 10 20 30 40 50 60 70 80 90 100
5.5	Were the parameters optimized to improve the process?	☐	☐	☐	0 10 20 30 40 50 60 70 80 90 100
5.6	Notes:	☐	☐	☐	0 10 20 30 40 50 60 70 80 90 100

Figure S 5: Exemplary questionnaire to determine technology maturity – stage 5 (adapted and translated from Schindler (2015)[1])

Questionnaire to determine the maturity of aerosol coating for the production of ceramic solid state cells

General information

Technology designation	Responsible person, function, organization/institute, department	Date
T2 Aerosol Coating		

6. Production structure

Maturity level description

Stage six comprises the integration of the technology into a production structure with upstream and downstream technologies. The performance of the technology and its interactions can be determined under real environmental conditions.

6.1
Has the aerosol coating been successfully integrated into a production line for the production of cell layers?

yes ☐ no ☐ uncertain ☐
Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

6.2
Is it possible to link up with upstream and downstream technologies taking into account the continuous technology chain?

yes ☐ no ☐ uncertain ☐
Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

6.3
Were the key figures and process parameters of the aerosol coating documented for the optimized production of a cell layer?

yes ☐ no ☐ uncertain ☐
Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

6.4
Has a functional electrolyte/electrode been produced with a technology chain that uses aerosol coating as a coating technology?

yes ☐ no ☐ uncertain ☐
Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

6.5
Is the performance of the aerosol coating for the production of the electrolyte/electrode under consideration/evaluation of the setpoint/actual value results given?

yes ☐ no ☐ uncertain ☐
Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

6.6
Notes:

yes ☐ no ☐ uncertain ☐
Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

Figure S 6: Exemplary questionnaire to determine technology maturity – stage 6 (adapted and translated from Schindler (2015)[1])

Questionnaire to determine the maturity of aerosol coating for the production of ceramic solid state cells

General information

Technology designation	Responsible person, function, organization/institute, department	Date
T2 Aerosol Coating		

7. Series application

Maturity level description

Technologies, which are assigned to the last maturity stage, the industrial usability is given. Only a minimal further development of the technology takes place. The focus is on standardization.

7.1

Has the aerosol coating been completely described by manufacturing and operating documents? (e.g. patents, certificates, guidelines, (DIN-) standards, etc.)

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

7.2

Is the aerosol coating already used in real production?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

7.3

Is the aerosol coating used to produce (oxide) ceramic coatings in real production?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

7.4

Is the aerosol coating used for the production of oxide ceramic solid state batteries in a real production?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

7.5

Is the know-how about the aerosol coating sufficiently secured? (Can every skilled employee of the company build up and use the technology with available documents?)
Is there a standardized process?

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

7.6

Notes:

yes ☐ no ☐ uncertain ☐

Degree of fulfilment / uncertainty: 0 10 20 30 40 50 60 70 80 90 100

Figure S 7: Exemplary questionnaire to determine technology maturity – stage 7 (adapted and translated from Schindler (2015)[1])

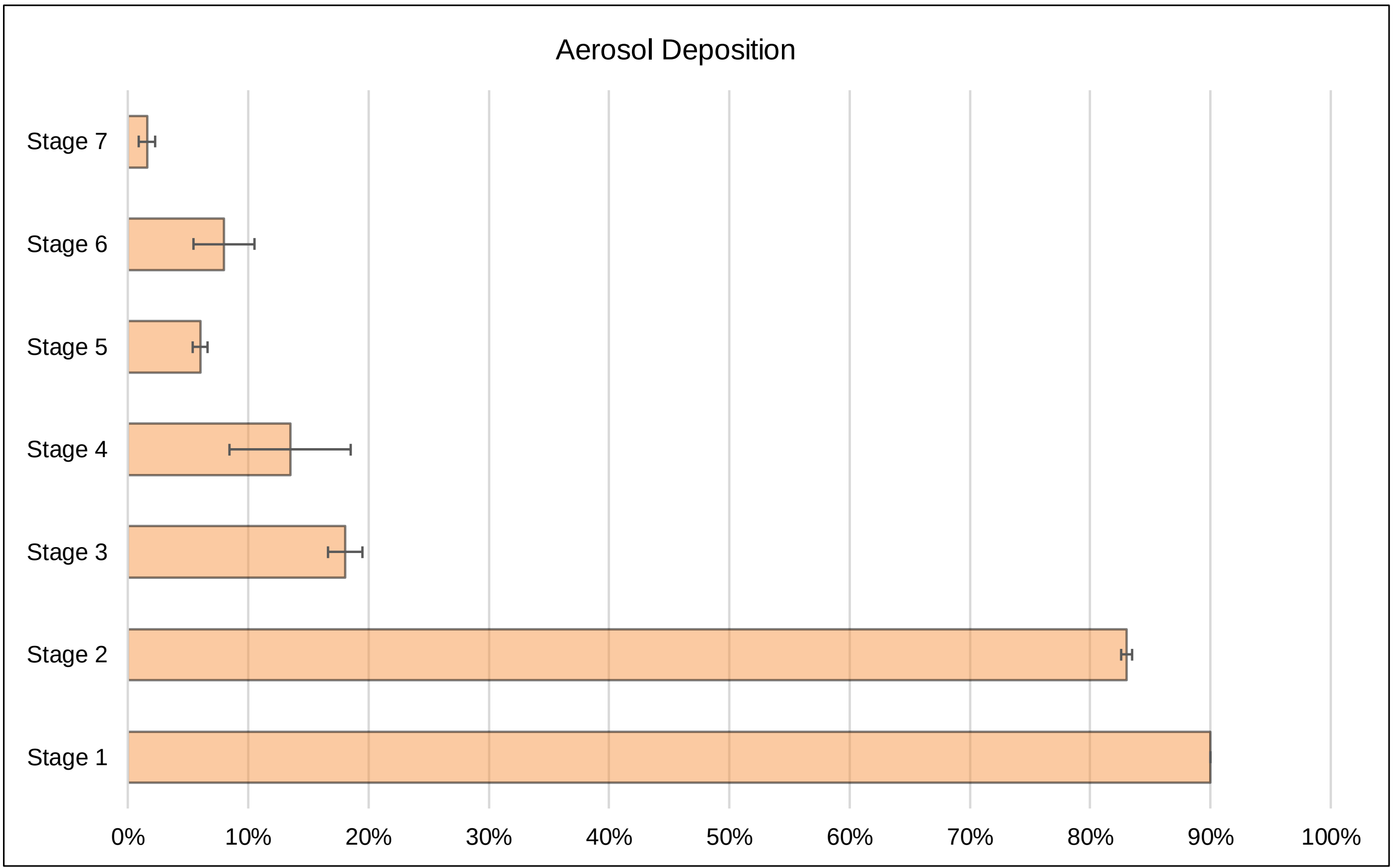

Figure S 8: Exemplary evaluation of technology readiness by stages 1-7

S3. Assumptions for cost estimation

Table S 5: Densities and prices of cell materials used for cost estimation

	Density	Ref.	**Cost**	Ref.
HE-NMC	4.25 g/cm³	2	20 $/kg	3
LNMO	4.4 g/cm³	4	21 $/kg	4
NMC 811	4.7 g/cm³	2	24 $/kg	3
Carbon Black	2 g/cm³	5	7.15 $/kg	6
PVdF	1.76 g/cm³	5	21.5 $/kg	7
LLZ	5.1 g/cm³	2	variable	
Li	0.534 g/cm³	8	250 $/kg	3
Al foil	2.7 g/cm³	8	6 $/kg	3
Cu foil*	8.92 g/cm³	8	13 $/kg	3
Al hard case (HEV)**	2.7 g/cm³	8	1.23 per housing	Calculated from Ref. [7]

*For the calculation, a bipolar Al current collector with a 100 nm thick Cu layer was assumed. Since no data for fabrication of such a thin Cu layer could be found in literature, the referenced price for Cu foil was assumed, which accounts to approximately twice the Cu raw material price (approx. 6 $/kg).
**The indicated reference states a price per PHEV2[9] hard case cell housing. Cost for a HEV cell housing were calculated by geometrical considerations, assuming that material cost and fabrication cost scale proportionally with dimensions.

Supplementary references

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