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Current status of a 1Ah class solid-state lithium metal secondary battery: Cycle and Rate capability

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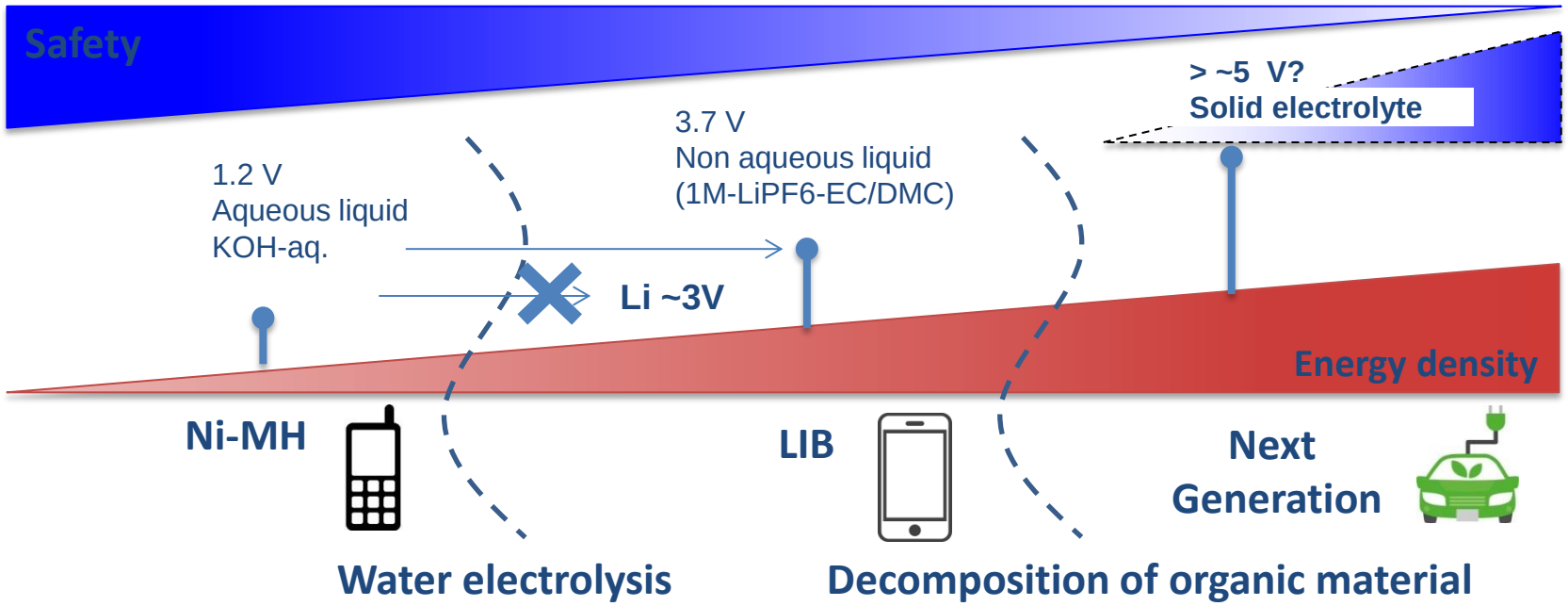
Samsung R&D Institute Japan (SRJ)

Beyond Lithium-Ion Workshop
Nice, Oct. 2nd 2018

Research motivation



- Desired properties -

- ✓ **Electrochemical stability**
- ✓ **Ideal Li^+ transport and reaction**
- ✓ **Safety (Strategy for Li-metal anode)**

Key characteristics in sulfide based ASSBs

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1. Softness of sulfide based electrolyte
2. High ionic conductivity >2 $mScm^{-1}$, $t^{+}=1$
3. Buffer layer on cathode active material

Large format cell

Issues:

- Process (compression/tact time)
- Supply chain
- Operation pressure
- Hydrolysis of sulfide materials

Mass Production

Softness of sulfide SEs

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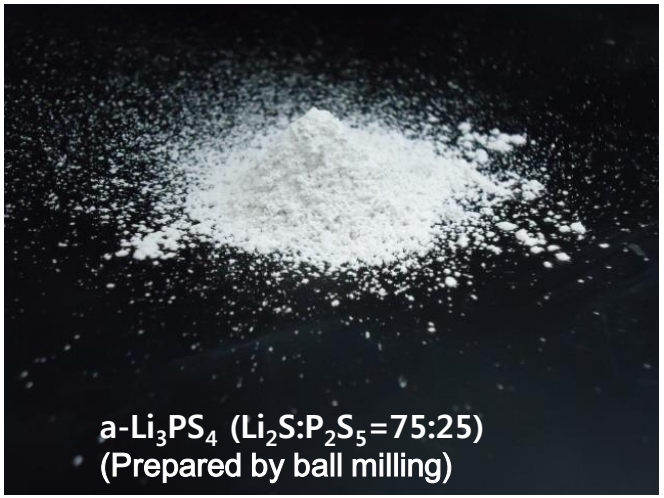
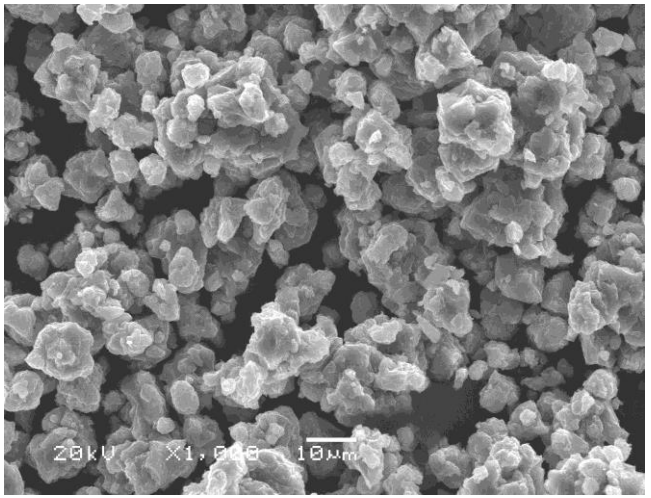
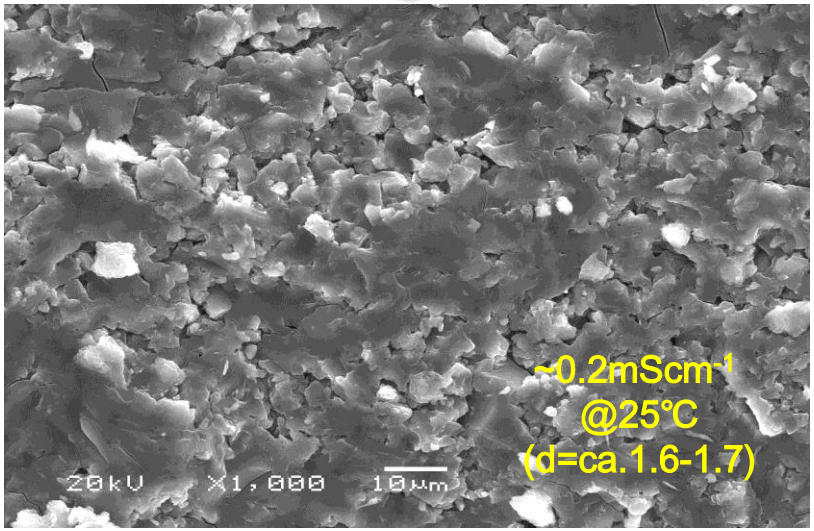




Cold press



Buffer layer

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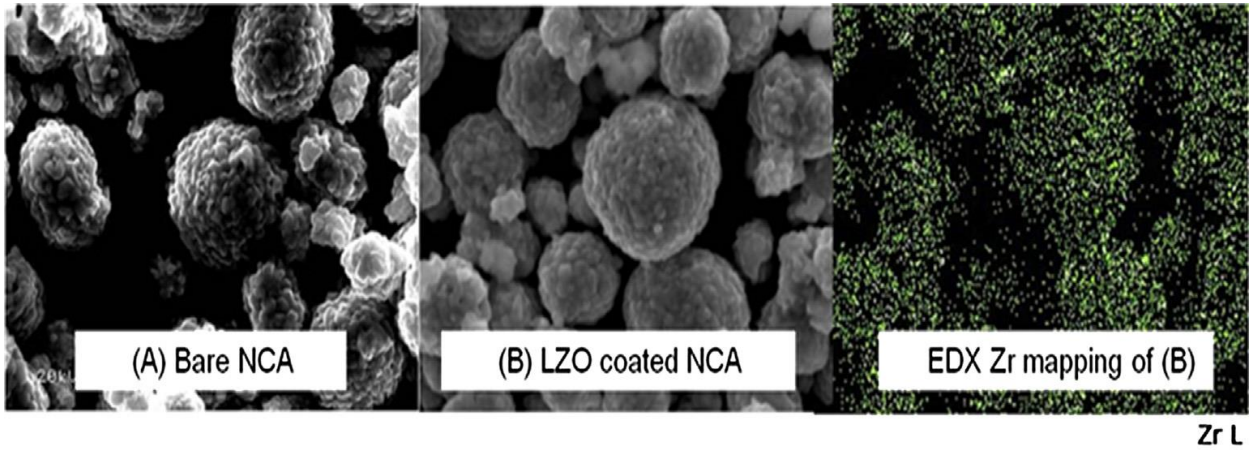
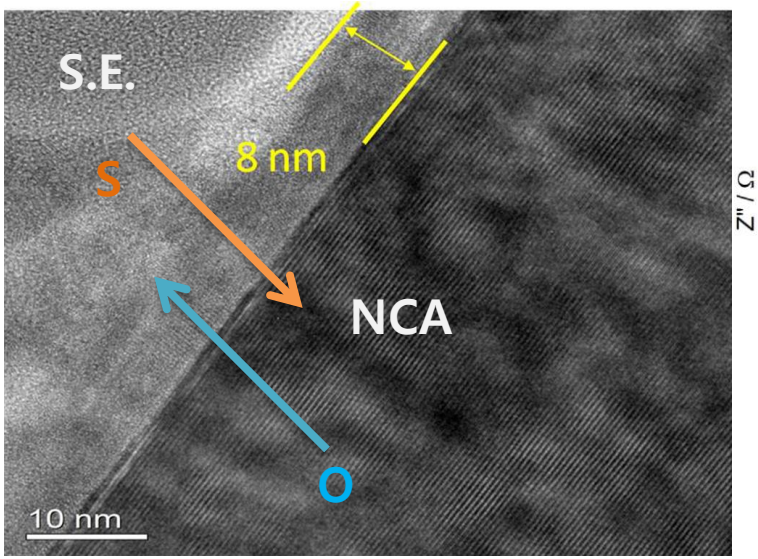
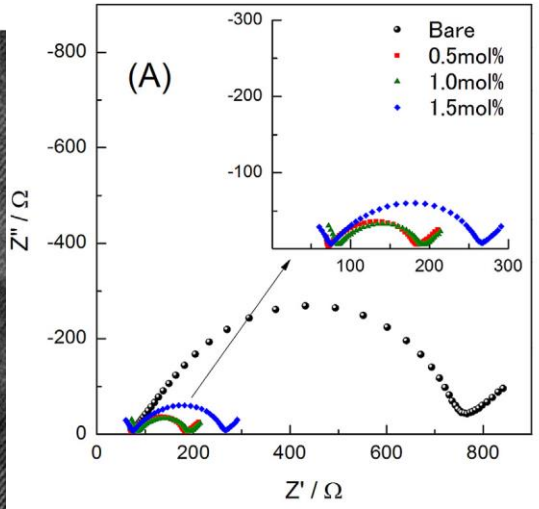
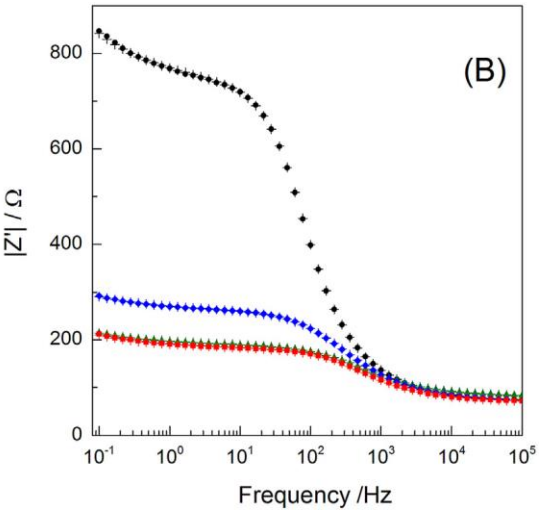
➢ Mutual diffusion must be prevented.



R_{CT} ↑ at high SOC







Ito et al., Journal of Power Source 248 (2014)943-950

R&D time line at Samsung

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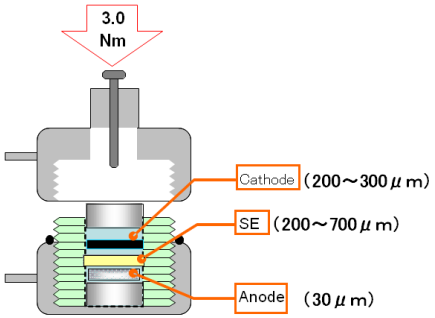

~2010 2014 2018

σ_{25}
$P_{(S.C.)}$
$N_{(S.C.)}$
Cell Q

1 Background

2 Pelletized cell: intrinsic characteristics

3 Prototype cell: current status

4 Conclusions

Preliminary test of a pelletized cell

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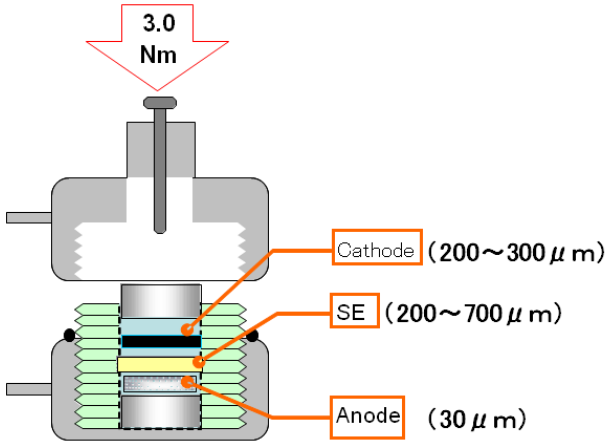
- Test cell for materials evaluation. (Neglect energy density.)
- No binder → Ideal performance

- 0.8~1.5 mAh
- Graphite, Si, Li/SE/LMeO, S
- Characterization of materials
- No binder (pellet)
- 13 mmΦ (1.33cm^2)



Cycleability

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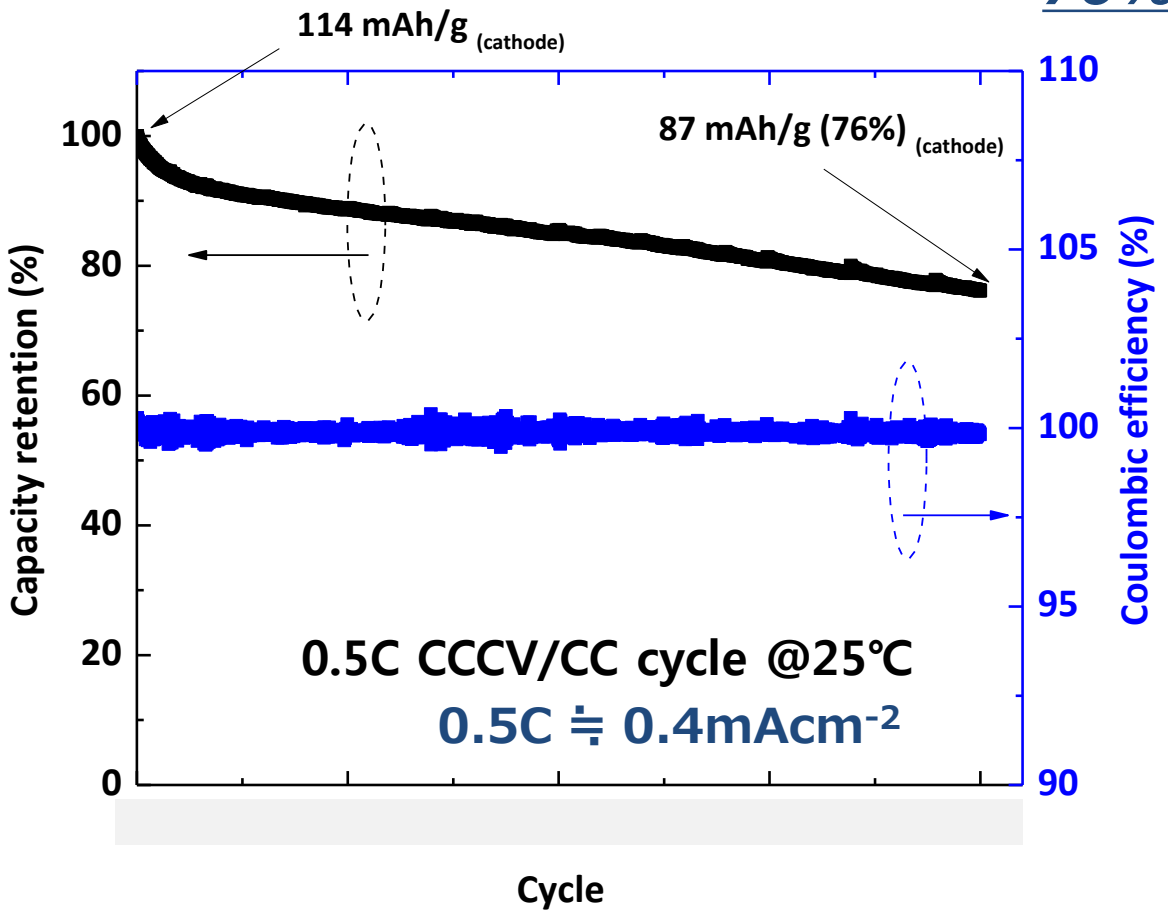
76% retention @2K cycle



Item	Spec.
Negative	Li foil t=30μm
Positive (wt%)	NCA:SE:C=60:35:5
Loading	0.83 mAh/cm^2
Cap. Ratio N/P	26
SE	LPSX
Cell capacity	1.1mAh (cathode)

- **Over 99.9% coulombic efficiency**
- **Negligible impact of reaction products**

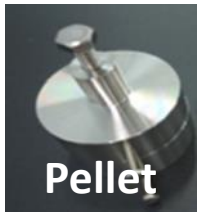
Rate capability

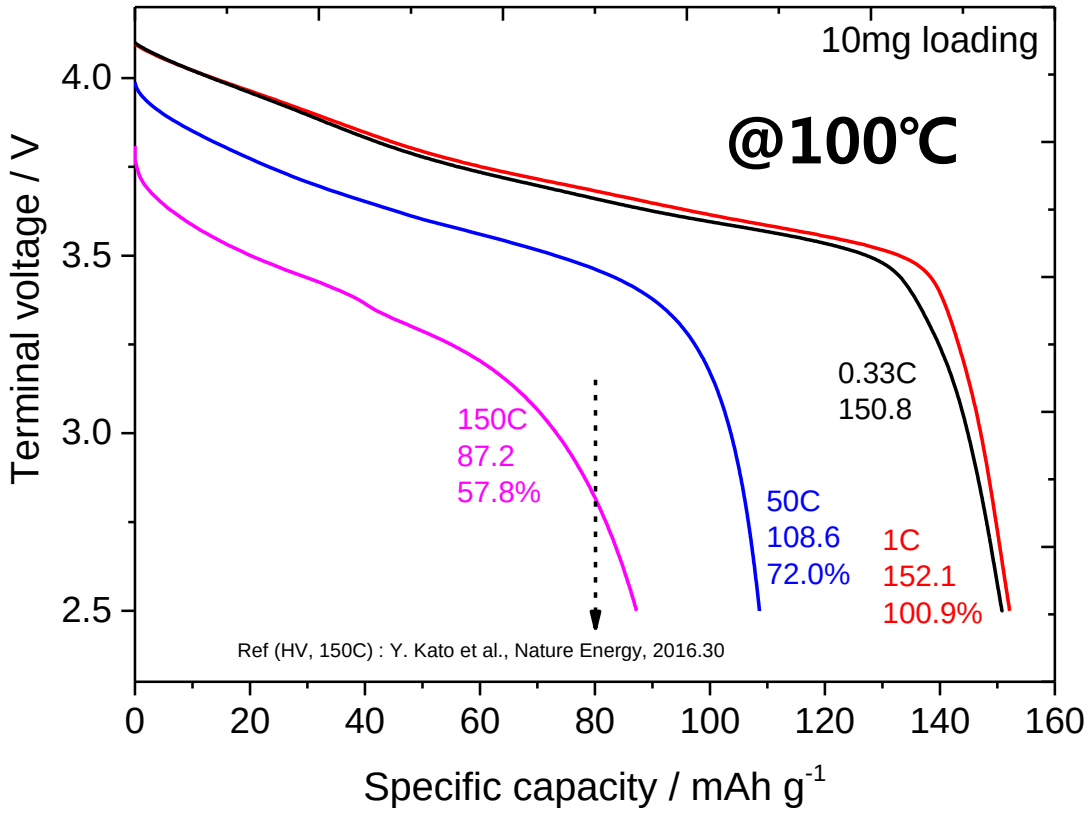
NCM: $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$

High rate discharge capability of a solid-state lithium secondary cell

13mmΦ pellet cell

Item	Spec.
Negative	Li foil t=30μm
Positive (wt%)	NCM:SE:C=60:35:5
Loading	0.6 mAh/cm^2
Cap. Ratio N/P	21
SE	$LiCl$-Li_2S-Li_3PS_4
Cell capacity	0.8 mAh (cathode)





➢ **58% discharge at 150C (24sec)**

@100℃

Y. Kato et al., ARTICLE NUMBER: 16030 | DOI: 10.1038/NENERGY.2016.30

A large gap : Pellet and Prototype cell

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- Continuous, large scale processes require wet process and binders.
- Unlike conventional LiBs, binders in ASSBs are insulator.
  → Significant reduction in electronic/ionic conductivity

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Solid State Ionics 158 (2003) 275–280

SOLID STATE IONICS
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Fabrications and properties of composite solid-state electrolytes

Taro Inada[a,*], Kazunori Takada[a], Akihisa Kajiyama[a], Masaru Kouguchi[a], Hideki Sasaki[a], Shigeo Kondo[a], Mamoru Watanabe[a], Masahiro Murayama[b], Ryoji Kanno[b]

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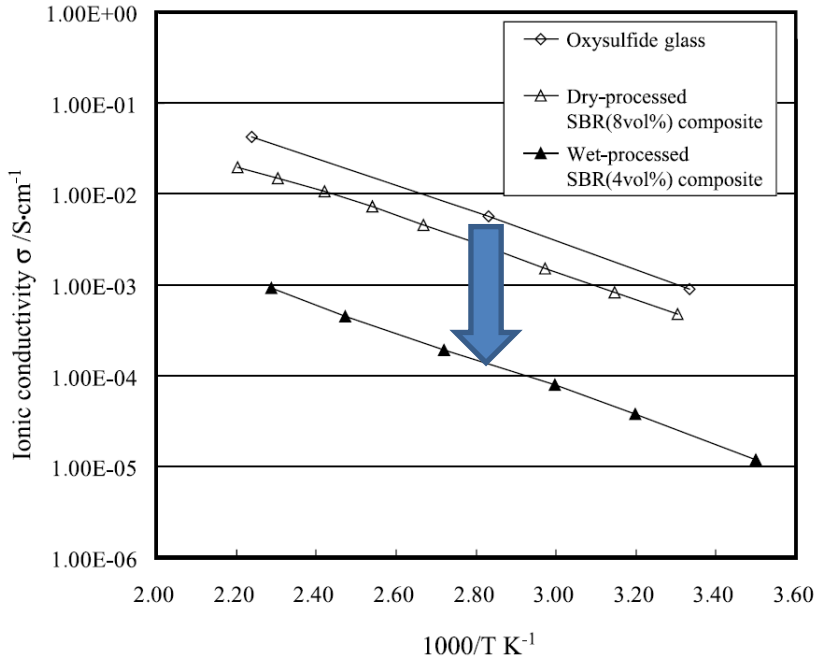

- Takada et al. propose dry process

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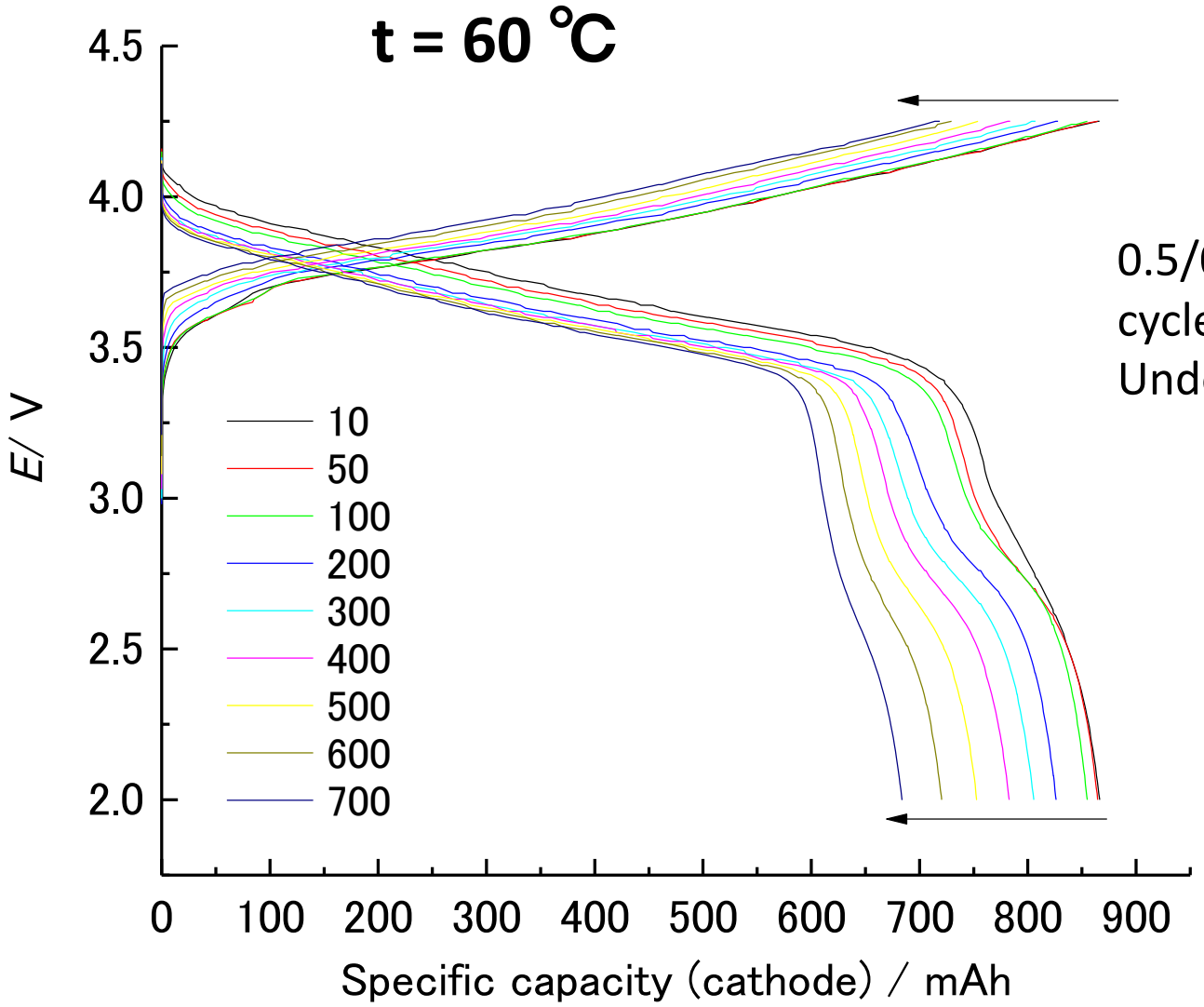
Cyclability



0.5/0.5C CC charge/discharge cycle @ 60℃
Under an external pressure.

2017 ver. Cell

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4 Conclusions

Conclusions

1: Sulfide solid electrolyte

- Applicable（Stable operation at Li potential. High voltage is more challenging.）
- 700 cycles、demonstrated 60℃ operation

2: Adaptation of Li-metal anode

- Excellent C.E. : No excess Li required
- Remaining issues :

  i) Need external pressure

  ii) Micro short circuit

3: Deposition-type Secondary ASSB

Very promising, but many outstanding issues and need to ensure safety

Acknowledgement

- Organization committees
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- SRJ/SAIT research staff