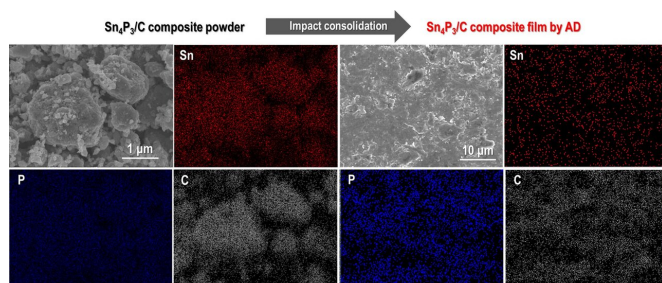


# Leap toward robust binder-less metal phosphide electrodes for Li-ion batteries

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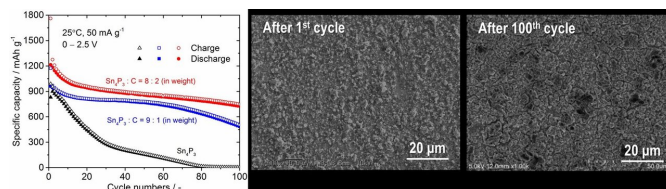
Scanning electron microscope (SEM) images of Sn<sub>4</sub>P<sub>3</sub>/C composite particles (1st row:1st image), and surface of Sn<sub>4</sub>P<sub>3</sub>/C composite film fabricated by the AD process (1st row:3rd image). Corresponding elementary distributions for Sn, P, and C are also shown. Credit: (c) Toyohashi University of Technology. All Rights Reserved.

Researchers at the Toyohashi University of Technology have successfully fabricated a binder-less tin phosphide (Sn<sub>4</sub>P<sub>3</sub>)/carbon (C) composite film electrode for lithium-ion batteries via aerosol deposition. The Sn<sub>4</sub>P<sub>3</sub>/C particles were directly solidified on a metal substrate via impact consolidation, without applying a binder. Charging and discharging cycling stabilities were improved by both complexed carbon and controlled electrical potential window for lithium extraction. This finding could help realize advanced lithium-ion batteries of higher capacity.

Lithium-ion (Li-ion) batteries are widely used as a power source in portable electronic devices. They have recently attracted considerable attention because of their potential to be employed on a large-scale as a power source for electric vehicles and plugin hybrid electric vehicles, and as stationary energy storage systems for renewable energy. To realize advanced Li-ion batteries with higher energy density, anode materials with higher capacity are required. Although a few Li alloys

such as Li-Si and Li-Sn, whose theoretical capacity is much higher than that of graphite (theoretical gravimetric capacity = 372 mAh/g), have been extensively studied, they generally result in poor cycling stability due to the large variation in volume during charging and discharging reactions.

Tin phosphide (Sn<sub>4</sub>P<sub>3</sub>, theoretical gravimetric capacity = 1255 mAh/g) with a layered structure, generally used as a high-capacity alloy-based anode material for Li-ion batteries, has an averaged operation potential of - 0.5 V vs. Li/Li<sup>+</sup>. Reports indicate that complexing [carbon](#) materials with nano-structured Sn<sub>4</sub>P<sub>3</sub> particles significantly enhance the cycling stability. Generally, electrodes used in batteries are fabricated by coating a slurry comprising [electrode](#) active materials, conductive carbon additives, and binders on metallic foils. For carbon complexed Sn<sub>4</sub>P<sub>3</sub> (Sn<sub>4</sub>P<sub>3</sub>/C) anodes (such as those reported in the literature), the weight fraction of the active materials in an electrode is decreased by approximately 60 - 70 % because of the use of significant quantities of conductive additives and binders to achieve stable cycling. Consequently, the gravimetric specific capacity per electrode weight (including those of conductive carbon additives and binders) is decreased significantly.



Cycling performance of Sn<sub>4</sub>P<sub>3</sub> and Sn<sub>4</sub>P<sub>3</sub>/C composite films for different carbon contents (left) in a two-electrode cell with Li metal as the counter electrode. Cycling test was performed at cell voltage windows ranging from 0 V to 2.5 V. SEM images of Sn<sub>4</sub>P<sub>3</sub>/C composite film surface after the 1<sup>st</sup> (middle) and 100<sup>th</sup> cycles (right) are also shown. Credit: (c) Toyohashi University of Technology.

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V-0.75 V (middle) and that at 0 V-1.25 V (right) are also shown. Credit: (c) Toyohashi University Of Technology.

Researchers at the Department of Electrical and Electronic Information Engineering, Toyohashi University of Technology, have successfully fabricated a binder-less  $\text{Sn}_4\text{P}_3/\text{C}$  composite film electrode for Li-ion battery anodes via aerosol deposition (AD). In this process, the  $\text{Sn}_4\text{P}_3$  particles are complexed with acetylene black using a simple ball-milling method; the obtained  $\text{Sn}_4\text{P}_3/\text{C}$  particles are then directly solidified on a metal substrate via impact consolidation without adding any other conductive additives or binders. This method enables enhancement of the fraction of  $\text{Sn}_4\text{P}_3$  in the composite to above 80%. Furthermore, structural change of the composite electrode is reduced and cycling stability is improved for both complexed carbon and controlled electrical potential window for lithium extraction reaction. The  $\text{Sn}_4\text{P}_3/\text{C}$  composite film fabricated by the AD process maintains gravimetric capacities of approximately 730  $\text{mAh g}^{-1}$ , 500  $\text{mAh g}^{-1}$ , and 400  $\text{mAh g}^{-1}$  at 100<sup>th</sup>, 200<sup>th</sup>, and 400<sup>th</sup> cycles, respectively.

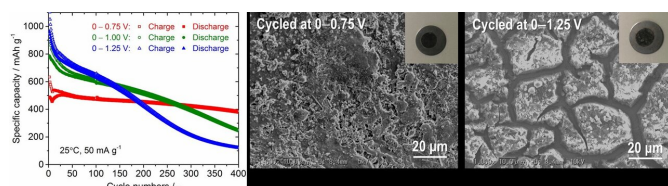
The first author Toki Moritaka is quoted as saying, "Although optimizing the deposition conditions was difficult, useful information on enhancement of cycling stability of the  $\text{Sn}_4\text{P}_3/\text{C}$  composite film electrode fabricated by the AD process was obtained. The complexed carbon functions not only as a buffer to suppress the collapse of electrodes caused by the large variation in volume of  $\text{Sn}_4\text{P}_3$ , but also as an electronic conduction path among the atomized active material particles in the composite."

"This process is an effective means to increase the capacity value per electrode weight. We believe there is scope for improvement of the electrochemical performance by the size and content of the carbon in  $\text{Sn}_4\text{P}_3/\text{C}$  used in composite film fabrication by the AD process. We are now trying to optimize the complexed carbon content and increase the composite film thickness," quotes Associate Professor Ryoji Inada.

The findings of this study may contribute to the realization of advanced Li-ion batteries of higher [capacity](#). Moreover, because not only Li but Na can also be stored in and extracted from  $\text{Sn}_4\text{P}_3$  by similar alloying and dealloying reactions, the  $\text{Sn}_4\text{P}_3$  electrode can be employed in next-generation Na-ion batteries at much lower costs.

**More information:** Toki Moritaka et al, Characterization of  $\text{Sn}_4\text{P}_3$ -Carbon Composite Films for Lithium-Ion Battery Anode Fabricated by Aerosol Deposition, *Nanomaterials* (2019). DOI: [10.3390/nano9071032](https://doi.org/10.3390/nano9071032)

Provided by Toyohashi University of Technology



Long-term cycling performance of  $\text{Sn}_4\text{P}_3/\text{C}$  composite films at different cell voltage windows ranging from 0 V to 0.75 V, and that from 1 V and to 1.25 V (left). SEM images of  $\text{Sn}_4\text{P}_3/\text{C}$  composite film surfaces cycled at 0

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