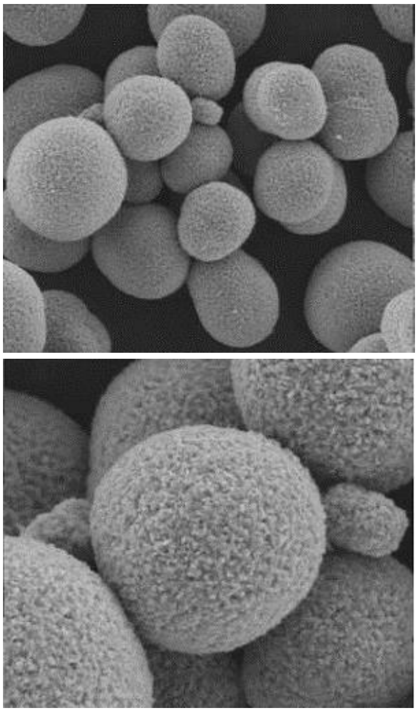

Taylor-Couette Crystallizer

EV Battery Materials

Business Case

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Taylor Vortex Reactor for EV Battery Materials

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Taylor Vortex Reactor for EV Battery Materials

Advanced Battery Materials for Sustainable Mass Production

LG Chem, a global leader in chemical manufacturing and R&D, as been using our pilot-scale 50L **Taylor Crystallizer** for advanced chemical research since 2012. LG Chem and its battery-focused subsidiary, LG Energy Solutions, are one of the most active companies in EV battery manufacturing, with partnerships with General Motors (LG Energy Solutions) and Stellantis (LGES). Our reactor is being explored as a key production technology to bring mass production of advanced battery materials to build domestic supply chains in the U.S.

Our Taylor-Couette Crystallizer is also the chosen technology used by researchers at **Argonne National Laboratory (ANL)**, the **MEET Institute** at the **University of Münster**, and the **ZSW Institute** in Germany.

Our proprietary LCTR Taylor crystallizer is the cornerstone of BAT167, a project commissioned by the U.S. Department of Energy’s Vehicle Technology Office for the development of **efficient, affordable EV battery technology**.

As stated in a recent review by the DOE, the Taylor Vortex Reactor is proving critical “to synthesize materials at a scale that bridges the needs of academia and industry-scale research and development (R&D).”

- 2021 VTO Annual Merit Review

Taylor Vortex Reactor for EV Battery Materials

Advanced Battery Materials for Sustainable Mass Production

Argonne National Laboratory’s public research compares the Taylor Vortex Reactor with batch tank reactors, continuous stirred tank reactors, and other conventional technologies, while providing advanced material samples to partnering research teams.

As such, the Taylor-Couette Crystallizer is contributing to the mission of replacing fuel-based vehicles with EV battery technology by 2050 *via* both commercial and R&D streams of production. This article will review the Taylor Vortex Reactor’s application and performance for the following competing technologies:

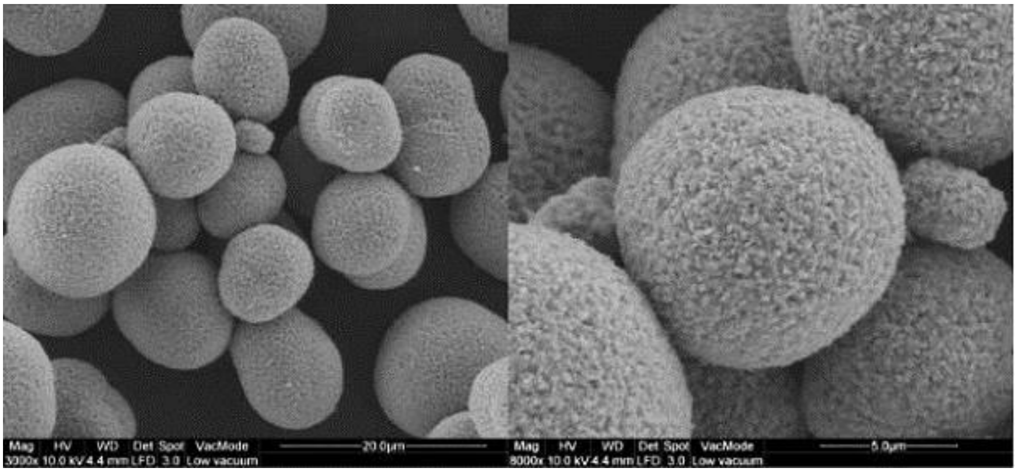
1. NMC Battery Materials
    1. Ni-Rich Hydroxide *via* Crystal Agglomeration
    2. High-Capacity Mn-Rich Cathode Materials
    3. Core Shel NMC Battery Material
2. Cobalt-free Lithium Battery Material
    1. Pilot-Scale Production of Spherical Co-Free Cathode Materials
    2. Co-free and Mn-free Cathodes for LIBs
3. NCA Hydroxide for Cathode Material
4. All Solid State NMC Battery Materials
    1. Ga-Doped LLZO Material
    2. Composite NCM-LZAOH Solid State Battery
5. Na-ion Battery Nanomaterials

Taylor Vortex Reactor for EV Battery Materials

Advanced Battery Materials Manufacturing

NMC Battery Materials, lithium-ion battery, Na-ion, solid state, and core-shell battery technology – the field of battery research is extremely diverse and expanding in the U.S. While several technologies display high electrochemical performance, common obstacles for battery R&D include high toxicity, high cost, and ethical sourcing concerns.

As a result, commercial and academic R&D industries are on a mission to develop battery technology that is high-performing, safer, more affordable, and more sustainable for mass production.



SEM images from Argonne National Laboratory's synthesis of NCM811 with the Taylor Vortex Reactor

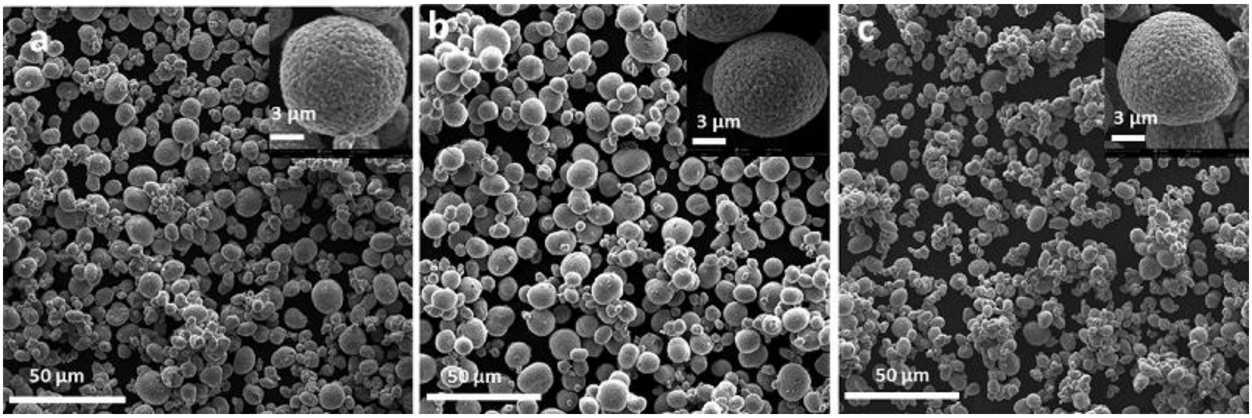
Taylor Vortex Reactor for EV Battery Materials

High-Performance Characteristics of Advanced Battery Materials

Next-Generation Battery Particles are often manufactured *via* co-precipitation or chemical crystallization. Qualitative characteristics indicative of high-quality battery particles include:

1) Spherical and uniform morphology, which lends the particles the desirable qualities of high tap density and structural integrity.
2) Narrow particle size distribution is also an indication of that electrochemical performance and structural integrity of the battery will be consistent and uniform across the battery cell.
3) Particle size to increase relative surface area. This is critical for powerful electrochemical performance, as the chemical reaction that produces energy occurs at the surface of the particles.

The precursor characteristics are heavily dependent on the crystal agglomeration mechanism, which depends on synthesis conditions, including reactor type and layout (Schmuch et al., 2020).

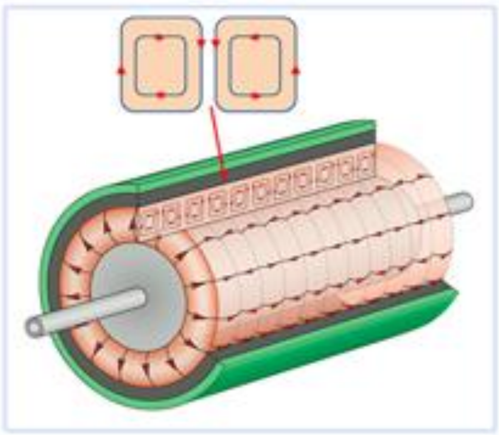


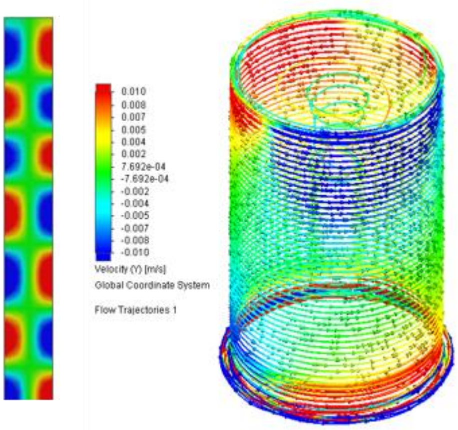
SEM images of (a) LNO, (b) LNMO, and (c) LNCO powder materials co-precipitated in Taylor Vortex Reactor (TVR) at MERF, Argonne National Laboratory

Taylor Vortex Reactor for EV Battery Materials

Taylor Vortex Reactor for Advanced Battery Materials

The Taylor Vortex Reactor (commonly referred to as Taylor Crystallizer, Taylor Flow Reactor, or Taylor-Couette Reactor) is a continuous-type flow reactor. It utilizes Taylor-Couette flow dynamics to uniform “micro-mixing zones” to create faster reaction kinetics and eliminate stagnant zones.





Read more on Taylor Crystallizer mechanism

The result is higher mass transfer, heat transfer, and fluid shear force compared to batch or continuous stirred tanks of comparable volumes. The Taylor crystallizer improves purity, morphology, particle size distribution, and crystallinity of particle products. Due to its continuous design and micro-mixing zones, the Taylor Crystallizer also displayed improved scalability from lab-bench to pilot to mass production scale projects.

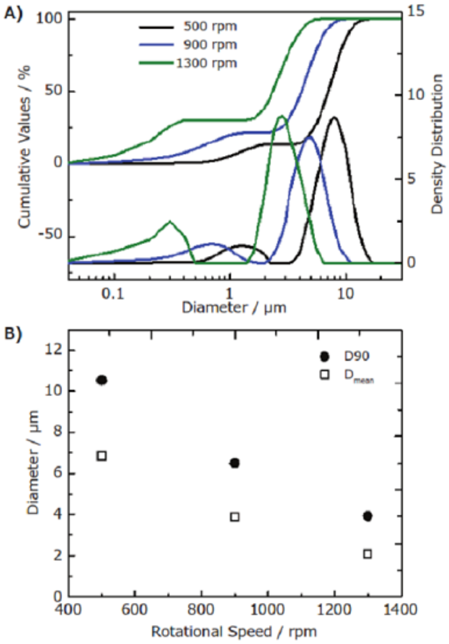
With options of pH control, temperature control, mean residence time, and varying hydrodynamic conditions *via* adjustable rotational speed, the Taylor Vortex Reactor creates nuanced possibilities for both R&D and mass production development for battery technology.

Taylor Vortex Reactor for EV Battery Materials

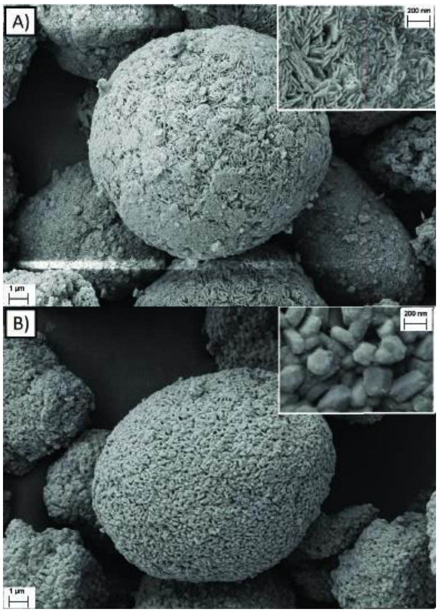
1. Taylor Crystallizer for NMC Battery Material for High-Energy LIBs

NMC Battery materials ($LiNi_xMn_yCo_zO_2$, where $x + y + z = 1$) are one of the largest categories of new lithium-ion battery materials.

MEET Institute's Dr. Richard Schmuch and colleagues' review of NCM811 provides a valuable overview of the Taylor Vortex Reactor for NCM materials. Specifically, they examine the continuous reactor's production of high-performing cathode materials for EV applications. In summary, they find that the TVR produces cathode materials with high densities, large particle size, bimodal size distribution, and spherical morphology.



(A) NMC811 cumulative values and density distribution produced by Taylor Vortex reactor. (B) Average D90 values and PSD of Taylor-made NMC battery materials



SEM images of (A) NMC Precursor Material produced by Taylor Vortex Reactor and (B) Lithiated NMC Material

Nickel-Rich Layered Cathode Materials for High-Energy Lithium Ion Batteries via a Couette-Taylor Flow Reactor. University of Münster, MEET Battery Research Center (2020)

Taylor Vortex Reactor for EV Battery Materials

1.1 Ni-Rich Hydroxide via Crystal Agglomeration

The production of Ni-rich NMC battery materials by D. K. Thai, Q.-P. Mayra and W.-S. Kim (2014) focuses on crystal agglomeration. It compares the performance of the Taylor Vortex Reactor with that of a continuous mixed-suspension mixed-product removal (MSMPR) crystallizer.

Crystal Agglomeration of NMC Hydroxide

Crystal agglomeration involves two consecutive processes:

1. Crystal aggregation

Aggregates form via the collision and physical binding of individual crystals caused by the velocity of the fluid motion. Simultaneously, the velocity of the fluid motion also creates a hydrodynamic fluid shear force that re-disperses the crystal aggregates.

2. Molecular Growth

The aggregates bind strongly enough to resist the hydrodynamic fluid shear force. This process is controlled by mass transfer velocity, whose effect on molecular growth is not linear.

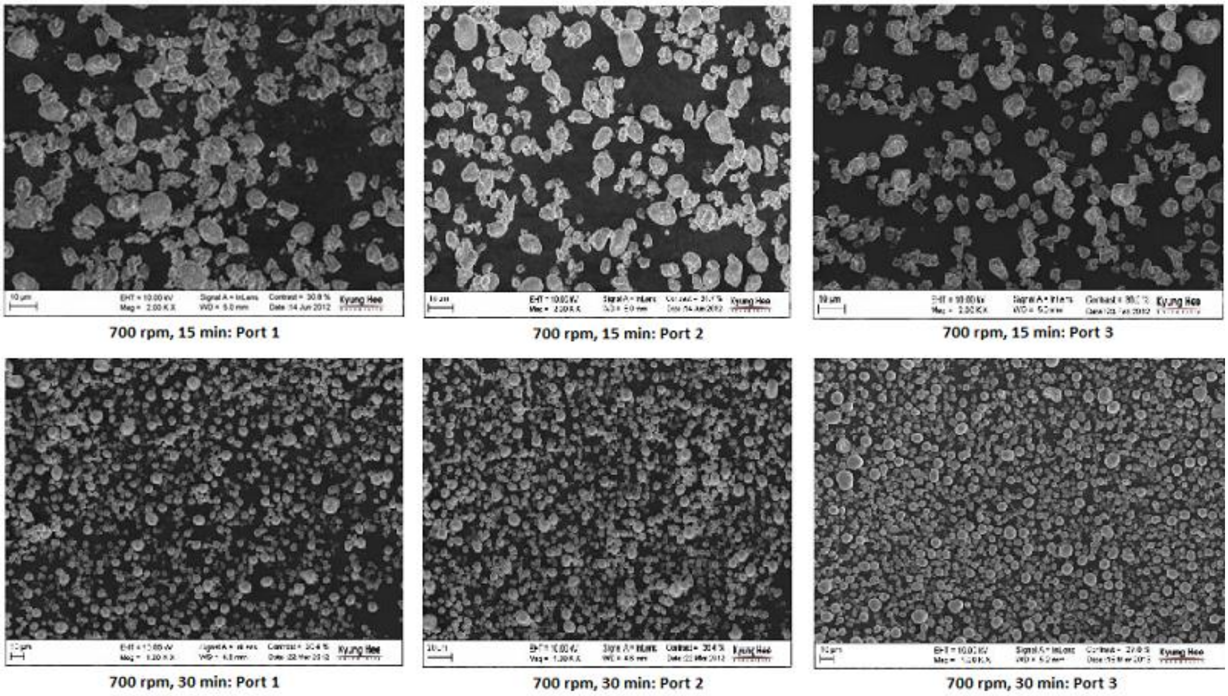
At the time of this study, it was already established that NMC batteries with a Ni-rich hydroxide precursor displayed high reversible electric capacity (>200 mAh/g). Specifically, spherical precursor particles with a narrow PSD have high packing density and surface area for efficient charge transfer. Precursor materials with these characteristics are demonstrated to have high electrochemical performance.

1.1 Ni-Rich Hydroxide via Crystal Agglomeration

Enabled to the Taylor Reactor’s easy operability, the researchers manipulated rotational speed, residence time, and pH to examine NMC hydroxide formation under various conditions. Additionally, Thai et al. (2015) compared crystal agglomeration with the Taylor Crystallizer’s various flow patterns (Taylor vortex flow, wavy vortex flow, modulated wavy flow, etc.) with the turbulent eddy flow of the MSMPR crystallizer.

NMC Battery Set-Up With Taylor Crystallizer

Thai et al.’s Taylor Crystallizer had three ports along axial direction the reactor. These ports allowed for sampling at different points of the crystallization process.


700 rpm, 15 min: Port 1
700 rpm, 15 min: Port 2
700 rpm, 15 min: Port 3
700 rpm, 30 min: Port 1
700 rpm, 30 min: Port 2
700 rpm, 30 min: Port 3

Morphology comparisons of NMC hydroxide samples from the Taylor Crystallizer

1.1 Ni-Rich Hydroxide via Crystal Agglomeration

Hydrodynamic Fluid Motion: The hydrodynamic conditions within the Taylor Crystallizer were controlled by adjusting the rotation speed of the inner cylinder from 300 rpm to 1500 rpm. The rotation speed is easily adjusted with a simple dial or via our touchscreen PLC system, which also displays other reaction parameters.

Mean Residence Time: The mean residence times of 5 min to 60 min were controlled by varying the flow rates of the feed solutions.

pH: A pH sensor was used to monitor the pH of the reaction, which was maintained between 10.0 and 13.0 by adjusting the flow rate of the NaOH solution. This adjustment can be done automatically by connecting the pH sensor to our solution pump.

NMC Battery Materials Produced Taylor Crystallizer

The Taylor Crystallizer produced NMC hydroxide particles with varying PSDs, depending on rotational speed and mean residence time. Thai et al. (2015) found that the most uniform and spherical particles were produced at a **rotational speed of 1500 rpm and a mean residence time of 30 min.**

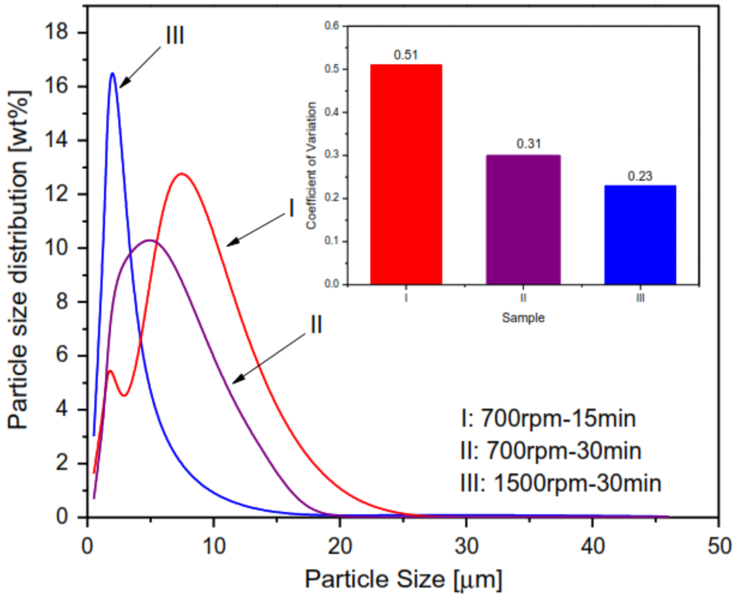
Particle morphology of Ni-rich hydroxide also varied according to reaction parameters, as summarized below.

Rotation Speed: 700 rpm
Mean Residence Time: 15 min

- First Sample Port: 15 µm with a PSD of 0.67 CV
- Outlet: 6 µm with a PSD of 0.51 CV

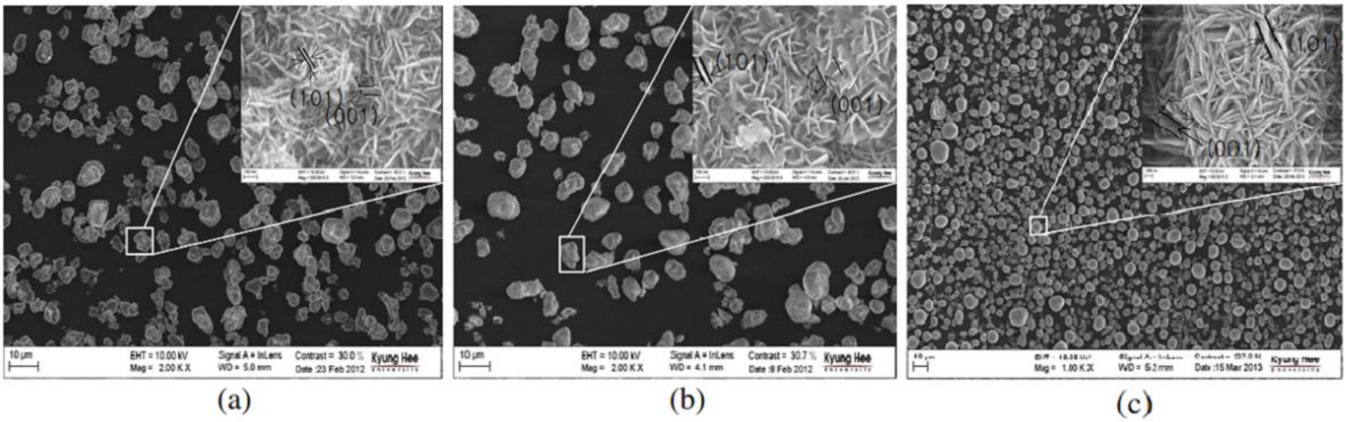
Rotation Speed: 1500 rpm
Mean Residence Time: 30 min

- First Sample Port: 4.5 µm
- Outlet: 3.6 µm



Taylor Vortex Reactor for EV Battery Materials

1.1 Ni-Rich Hydroxide via Crystal Agglomeration



Morphology of NMC Hydroxide Produced in Couette-Taylor Crystallizer at (a) 700rpm 15min (b) 700rpm 30min (c) 1500rpm 30min

Generally, particle size distribution and particle shape are key factors determining tap-density of the NMC battery precursors. As the Ni-rich hydroxide particles spent more time in the Taylor Crystallizer, they were exposed to fluid shear forces for extended periods. As a result, particles became more spherical which, in turn, increased tap density.

The particles produced at 1500 rpm with a mean residence time of 60 min had a tap density of 2.13 g/cm3, the highest of any previously reported values.

Thai et al. (2015) also demonstrated the importance of pH in the formation of Ni-rich hydroxide particles. By controlling the pH of the reaction by adjusting NaOH flow rate, the effects of pH 10 to 13 were observed for NMC hydroxide particle formation. In the present study, they found that optimal Ni-rich hydroxide formation occurs at a pH of 12.0.

Taylor Crystallizer vs. MSMPR Crystallizer for NMC Battery Materials

Thai et al. (2015) compared the performance of a Taylor Crystallizer (AKA Taylor Vortex Reactor) with that of a Rushton-type MSMPR crystallizer.

Reaction parameters were kept at the following constants:

- Rotation / agitation speed: 1200 rpm
- pH: 12.0
- Ammonia concentration: 15.0 mol/L

Mean residence time varied as follows:

- **Taylor Crystallizer: 30 to 60 min**
- MSMPR Crystallizer: 270 to 720 min

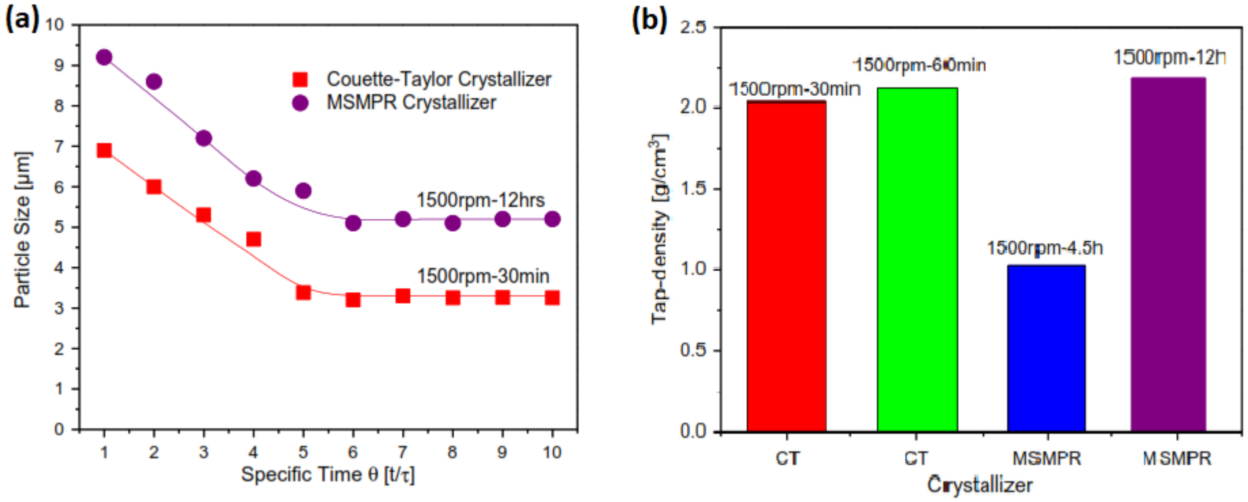
NMC hydroxide particles produced with a mean residence time of 30 min in the Taylor Crystallizer were 3.2 µm in size. In comparison, the MSMPR Crystallizer only produced particles of 5.2 µm in 12 h. This is due to the drastically higher hydrodynamic fluid shear of the Taylor Crystallizer.

As mentioned above, hydrodynamic fluid shear is a critical force in producing spherical and uniformly sized particles. When comparing viscous energy dissipation, **the Taylor Crystallizer displayed equivalent fluid shear at 300 rpm as the MRMPR’s random turbulent flow at >800 rpm.**

Taylor Vortex Reactor for EV Battery Materials

1.1 Ni-Rich Hydroxide via Crystal Agglomeration

While the MSMPR’s maximum tap density was higher than the Taylor Crystallizer, the drastically shorter residence time of the latter makes it more productive and practical for Ni-rich hydroxide production.



Comparison of Ni-rich hydroxide production between Taylor-Couette Crystallizer and MSMPR Crystallizer at steady state. (a) Transient profiles and (b) tap density of particles.

1.2 High-Capacity Mn-Rich Cathode Materials

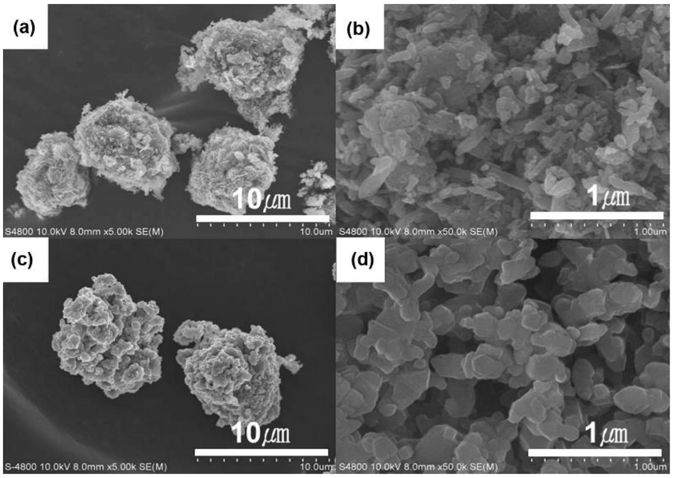
LiCoO2 is an important commercial material due to its high electrochemical performance. However, both lithium and cobalt face challenges of high cost, toxicity, safety, and issues concerning ethical sourcing. To explore cheaper and safer alternatives for high-capacity cathode materials, M. Choi et al. (2014) synthesized NMC rich in manganese.

NMC battery materials co-precipitation in a Taylor crystallizer:

- Materials sourcing: Daejung Co., Aldrich Korea, J.T. Baker
- **Mean residence time: 50 min**
- Rotation speed: 1500 rpm

Narrow PSD was observed with total residence times of from 150 min to 750 min residence times. With increasing reaction time, increasing mean particle size and narrowing PSD were observed.

- Spherical morphology, 7-8 µm in diameter (FE-SEM)
- Secondary particles formed by agglomeration of smaller particles <200 nm in diameter



FE-SEM images of Taylor-synthesized Mn-rich NMC precursor (a, b) and lithiated Mn-rich cathode material (c, d).

Taylor Vortex Reactor for EV Battery Materials

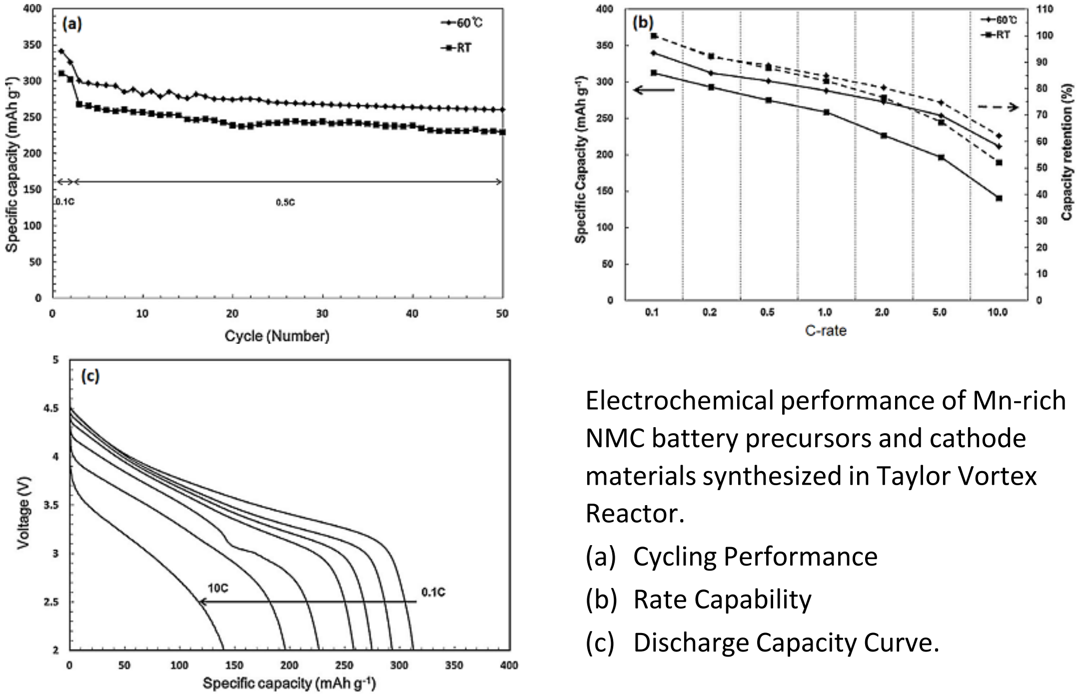
1.2 High-Capacity Mn-Rich Cathode Materials

Spherical cathode particles with rough surfaces were synthesized with a shorter reaction time in the Taylor-Couette reactor. The NMC cathode material is expected to demonstrate good high-rate electrochemical performance due to good absorption of the electrolyte.

- XRD Patterns: well-defined hexagonal structure, superlattice ordering of Li and Mn in transition-metal layers, uncontaminated lithium layer
- ICP Analysis: near-theoretical chemical compositions of NMC hydroxide precursor and Li-NMC cathode material

Coin-type cells were prepared with the Mn-rich NMC battery materials and tested for electrochemical performance. Discharge capacities:

- First charging capacity at 110 mAh/g: 4.45 V
- Corresponds with theoretical capacities associated with oxidation of Ni and Co ions.



Electrochemical performance of Mn-rich NMC battery precursors and cathode materials synthesized in Taylor Vortex Reactor.

(a) Cycling Performance

(b) Rate Capability

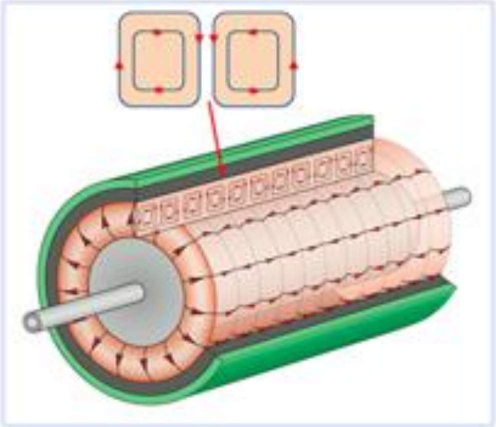
(c) Discharge Capacity Curve.

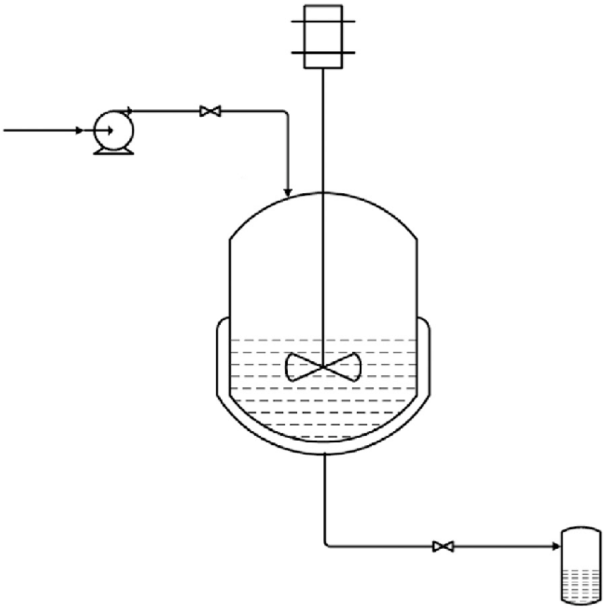
Taylor Vortex Reactor for EV Battery Materials

1.3 Core Shell NMC Battery Material

The Taylor Crystallizer has also been used to successfully produce uniform and spherical core-shell particles for high-performing NMC battery materials by Kim & Kim (2018). Core particles and shell particles were separately synthesized in the Couette-Taylor Crystallizer. Core and shell particles were simultaneously fed into the Taylor Reactor and made to adhere via collision and agglomeration within the reactor.

Core-shell NMC production with the Taylor Crystallizer was compared with an MSMPR Crystallizer.





Taylor Vortex Reactor for EV Battery Materials

1.3 Core Shell NMC Battery Material

Experimental Set-Up of Couette-Taylor Core Shells

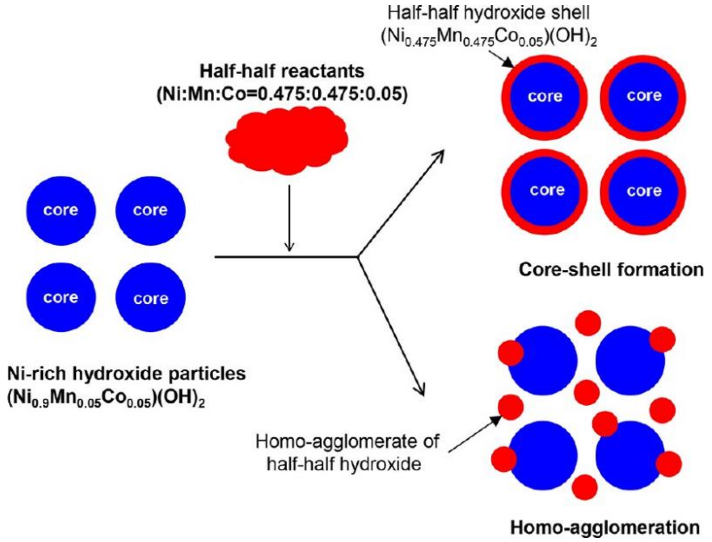
The Ni-rich NMC core particles were prepared via continuous precipitation in a Taylor Vortex Reactor with a Ni:Mn:Co ratio of 90:5:5.

- Temperature was maintained at 50°C with regulating jackets
- Mean residence time of 60 min
- Rotational speed of 1500 RPM
- pH was maintained at 12.5 by the automatic adjustments made to the flow rate of the NaOH solution upon feedback from the crystallizer’s pH sensor.

The Ni-Mn “half-half” shell particles were also prepared in the Couette-Taylor Crystallizer, at a Ni:Mn:Co ratio of 47.5:47.5:5.

These compositions maximized the advantages of Ni, Mn, Co while minimizing the high cost associated with cobalt. High nickel and cobalt content increases electrochemical performance, while high manganese increases stability.

The Ni-rich NMC core was structured for high electrochemical performance, while the half-half Ni-Mn shells was structured for high structural and thermal stability.



1.3 Core Shell NMC Battery Material

Core-Shell Particles

The purpose of the shell layer is to protect core particles from the toxic environment created within a battery cell. Thickness and uniformity of the shell layer are critical determinants of the properties of the core-shell particle. If the shell layer is too thin, it is unable to stabilize the core particle. However, if the shell layer is too thick, it will lower the electrochemical capacity of the core.

Moreover, highly spherical and uniform core particles are crucial in increase packing density, which increases overall electrical capacity of the battery cathode.

These critical characteristics are determined by the precipitation process of the core and shell particles. Therefore, the quality and productivity of the crystallizer are of utmost importance in producing high-quality, scalable core-shell NMC battery materials.

Core-Shell Synthesis via Collision and Agglomeration

Once the core and shell particles were precipitated, they were simultaneously fed into the Couette-Taylor Crystallizer for collision and agglomeration processes.

- Mean residence time varied from 20 to 120 min by adjusting the flow rates of the feed solutions
- Rotational speed varied from 300 to 1500 RPM to adjust the hydrodynamic conditions within the reactor
- pH varied from 9.5 to 11 by adjusting the flow rate of the NaOH reactant solution.

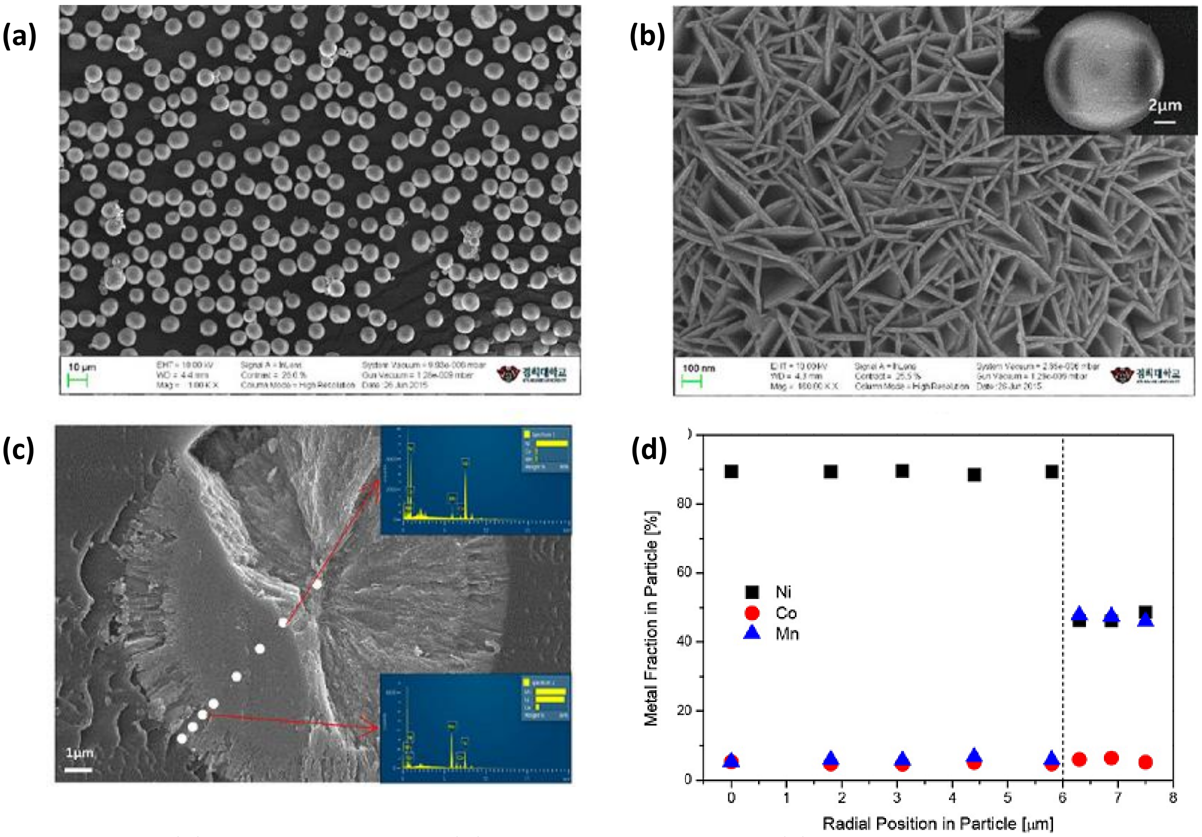
The MSMRP Crystallizer was a standard Rushton-type mixing tank with four baffles, a three-blade impeller, and a heating jacket on the wall of the reactor.7

Taylor Vortex Reactor for EV Battery Materials

1.3 Core Shell NMC Battery Material

Results of Core-Shell NMC Battery Synthesis in Couette-Taylor Crystallizer

- Morphology: spherical, uniform
- Mean particle size: 12.5 µm with 0.15 CV
- Tap density: 2.26 g/mL *The best value represented in literature at the time
- Energy dispersive spectroscopy (EDS) microscopic cross-section: 1-2 µm uniform shell layer
- Experimental confirmation of theoretical compositions: Ni-Mn half-half hydroxide shells and Ni-rich NMC core particles.



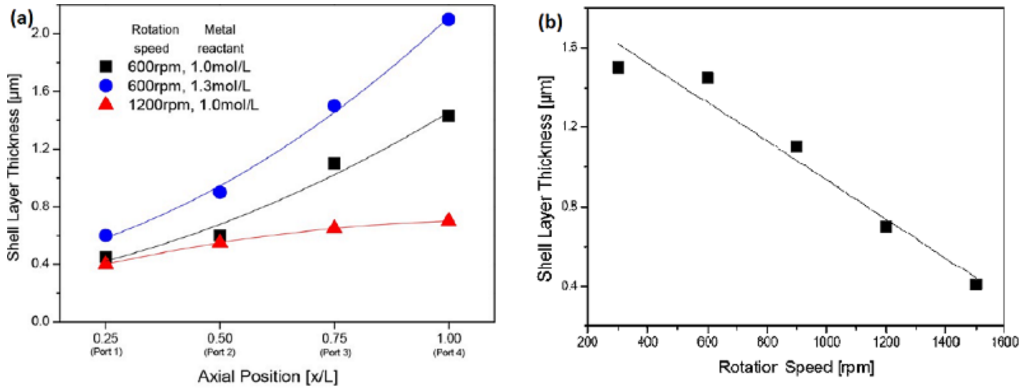
Morphology of (a) core particle shape, (b) surface of core particle. (c) EDS cross-sectional image of 1-2 µm thick shell layer (c) experimental metal compositions of core and shell reflect theoretical values.

Sample Outlet Ports & Varying Hydrodynamic Conditions

Moreover, the researchers’ Taylor Vortex Reactor was configured with multiple outlet ports along the length of the reactor. This allowed Kim & Kim (2018) to sample materials throughout the agglomeration process.

They found that the shell thickness increased monotonically along the length of the Couette-Taylor Crystallizer. Shell thickness also increased when the rotational speed was reduced and when the shell solution concentration increased. In other words, **shell layer formation depended heavily on the adjustable operating parameters of the Couette-Taylor Crystallizer, the reactant concentration, and the residence time**.

By adjusting the rotational speed of the inner cylinder, Kim & Kim (2018) generated different hydrodynamic conditions with the Taylor Crystallizer. This allowed them to observe the effect of different fluid motions on shell layer formation. Based on their quantitative observations, they concluded that the collision of primary-core particles is directly dictated by the fluid motion dominating the reaction.



Influence of Taylor Crystallizer operational parameters, reactant concentration, and residence time on thickness of NMC shell layer.

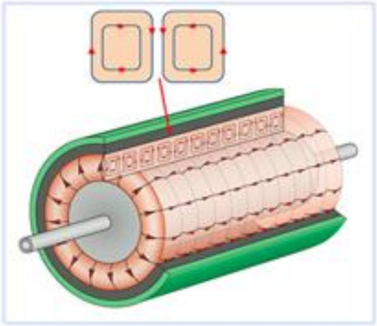
1.3 Core Shell NMC Battery Material

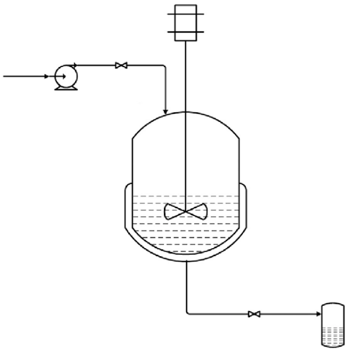
NMC Core-Shell Synthesis: Taylor Crystallizer vs. MSMPR Crystallizer

Kim & Kim (2018) compared core shell synthesis in a Taylor Crystallizer to an MSMPR Crystallizer. The Taylor Crystallizer generated variations of the Taylor flow as the dominant hydrodynamic conditions. In contrast, the MSMPR generated a random turbulent eddy with a 6-blade impeller.

A mean residence time of over 8 hours was required to form the desired core-shell particles in the MSMPR. When mean residence time was increased to 12 hours, both the thickness of the shell layer and the tap density of the core-shell particles.

Taylor Crystallizer	MSMPR Crystallizer
Mean Residence Time: 1 h	Mean Residence Time: 8 h
Shell layer thickness: 1.4 µm	Shell layer thickness: 1.3 µm
Tap density: roughly 2.04 g/cm^3	Tap density: 2.0 g/cm^3
Mean Residence Time: 1.5 h	Mean Residence Time: 12 h
Shell layer thickness: 2.1 µm	Shell layer thickness 1.6 µm
Tap density: roughly 2.06 g/cm^3	Tap density: 2.15 g/cm^3



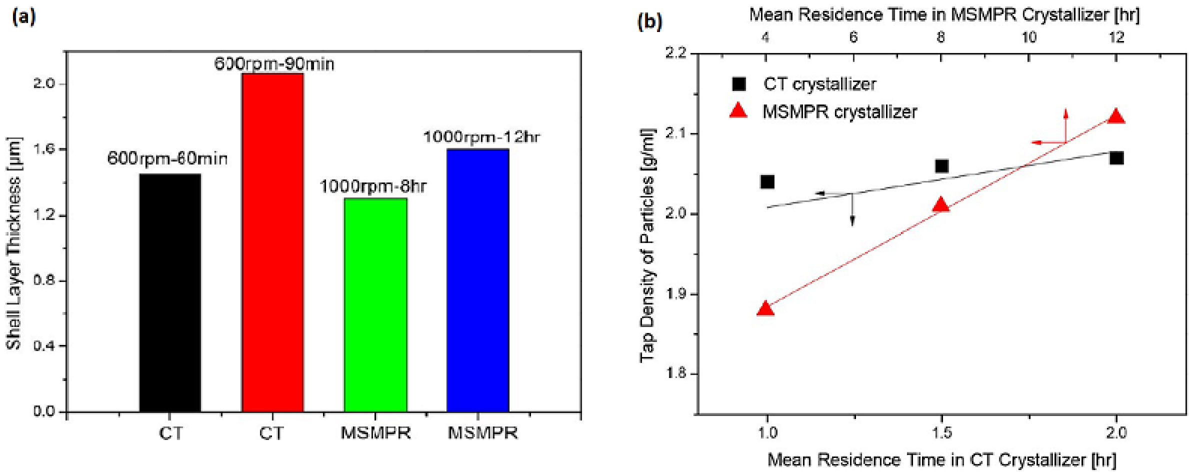


Taylor Vortex Reactor for EV Battery Materials

1.3 Core Shell NMC Battery Material

While the core-shell characteristics were desirable, synthesis in a MSMRP Crystallizer required significantly longer reaction time than in a Couette-Taylor Crystallizer.

Therefore, the researchers concluded that the Taylor-Couette Crystallizer is 10 times more efficient in producing NMC core-shell particles than a conventional MSMPR Crystallizer due to the Taylor vortex fluid motion.



NMC Core-Shell Crystallization in a Taylor-Couette Reactor vs. MSMPR (a) Shell Layer Thickness vs. Mean Residence Time, (b) Tap Density vs. Mean Residence Time.

Taylor Vortex Reactor for EV Battery Materials

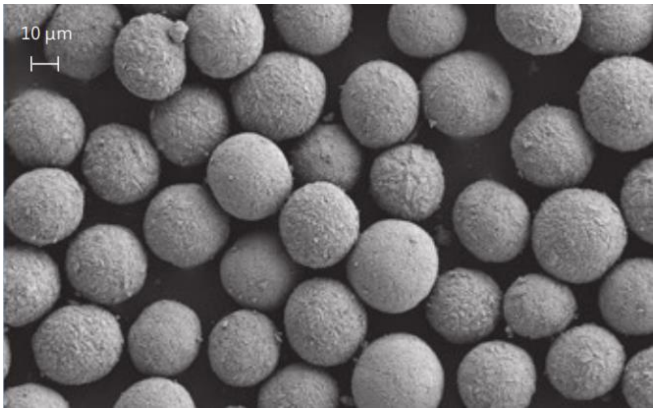
2. Cobalt-Free Cathode Material

2.1 Pilot-Scale Production of Spherical Cobalt-Free Cathode Powder

In partnership with the German Federal Government, ZSW has been developing next-generation lithium-ion battery materials with our Taylor Vortex Reactor. Specifically, they are developing cobalt-free cathode materials to increase specific energy, or energy density, while cutting costs of lithium batteries.

With our **1L Taylor Vortex Reactor**, ZSW synthesizes spherical cathode powders in the scale of 10 kg to 30 kg for electrode coating and cell assembly.

Excerpt of the ZSW Annual Battery Report 2020



Right: Cobalt-free cathode material produced with TVR at ZSW.

Left: ZSW researcher seen operating 1L Taylor Vortex Reactor

2.2 Cobalt-Free and Manganese-Free Cathodes for Lithium-Ion Battery

In a study by S. Aryal et al. (2021), the **Materials Engineering Research Facility (MERF) at Argonne National Laboratory** utilized our 1L Taylor Vortex Reactor (TVR) to study the effect of Mn and Co on Ni-rich layered cathode materials. Our Taylor Vortex Reactor was specifically selected for its scalability from R&D to large-scale production. In contrast, a small batch reactor is difficult to scale up for large-scale production.

Aryal et al. (2021) compared the performance of NMC, Co-free, and Mn-free cathode materials, which were all co-precipitated in the Taylor Vortex Reactor:

- $Ni_{0.9}Mn_{0.1}(OH)_2$
- $Ni_{0.9}Co_{0.1}(OH)_2$
- $Ni(OH)_2$

MERF's study was the first to published experiment in which the three compositions were synthesized via the same method in a Taylor Vortex Reactor and studied under identical conditions.

Taylor Crystallizer Set-Up

- Metal sulfate solutions, NH_4OH (ammonia) and NaOH (sodium hydroxide) were continuously fed into the reactor via 3 separate pumps
- The pH of the solution was maintained between 11 and 12
    - pH sensor was connected to the NaOH pump and automatically adjusted the flow rate to maintain desired pH
- Temperature was fixed at 50°C and monitored by a temperature sensor
- Rotational speed was maintained at roughly 800 RPM

Taylor Vortex Reactor for EV Battery Materials

2.2 Cobalt-Free and Manganese-Free Cathodes for Lithium-Ion Battery

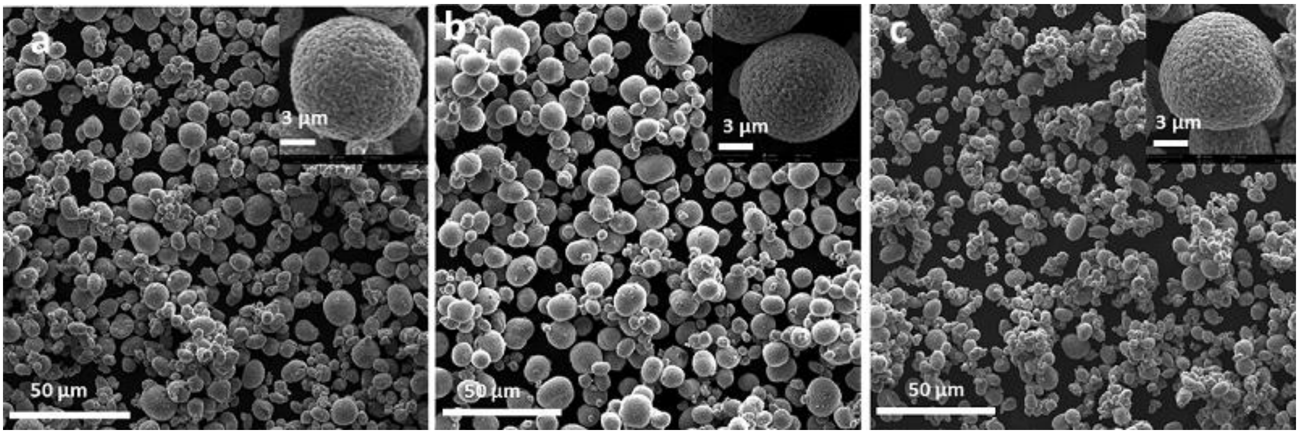
Mn-free and Co-free Battery Materials with Taylor Vortex Reactor

The Ni-rich battery materials synthesized in the Taylor Vortex Reactor were similar in size and spherical surface morphology. Primary particles of 200 nm – 300 nm aggregated to form secondary particles with an average size of 10-15 µm.

Tap Density:

- LNO: 1.99 g/cc
- LNMO: 2.14 g/cc
- LNCO 1.82 g/cc

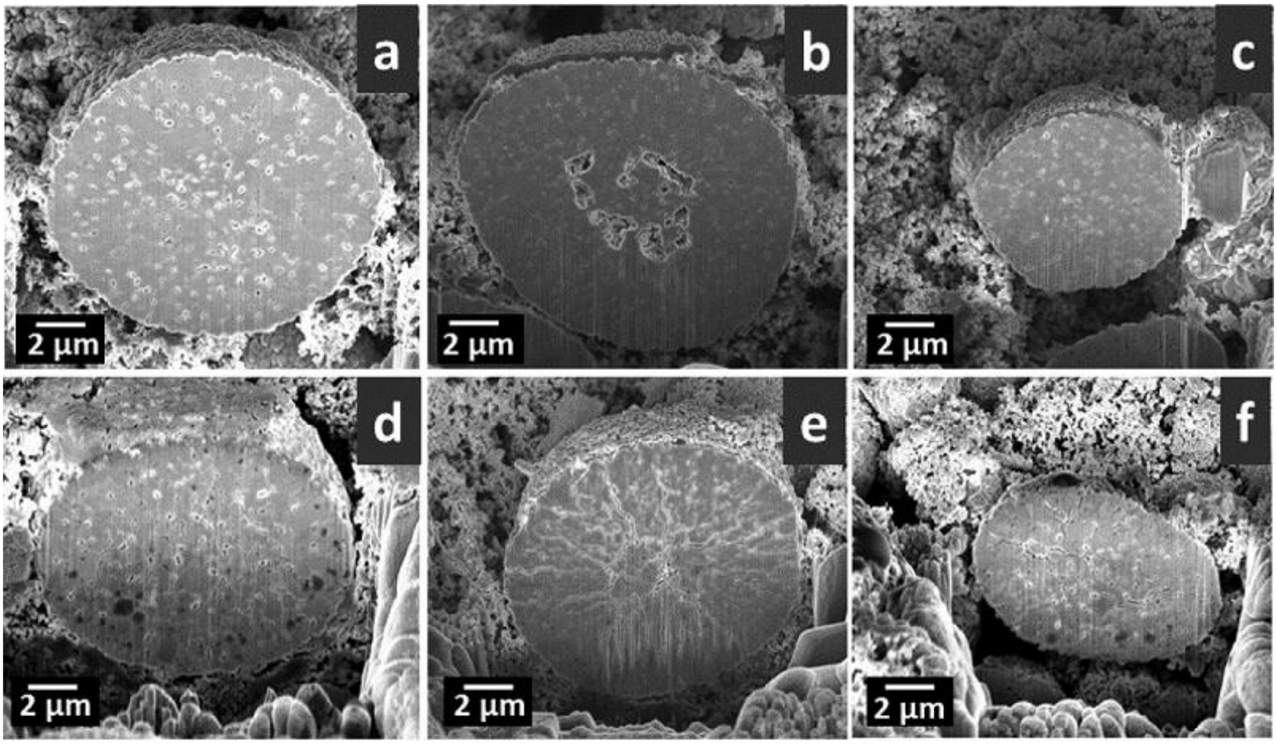
The high tap density of LNMO is due to individually denser particles, as well as a narrower particle size distribution, which allows for denser packing.



SEM images of (a) LNO, (b) LNMO, and (c) LNCO powder materials co-precipitated in Taylor Vortex Reactor (TVR) at MERF, Argonne National Laboratory.

Taylor Vortex Reactor for EV Battery Materials

2.2 Cobalt-Free and Manganese-Free Cathodes for Lithium-Ion Battery



SEM Images of pristine and cycled (a, d) LNO, (b, e) LNMO, and (c, f) LNCO cathode particles synthesized in Taylor Crystallizer.

In conclusion, MERF researchers found that Co-free LNMO cathode material demonstrates highly stable cyclic performance and is a noteworthy contender for sustainable next-generation Li-ion battery material.

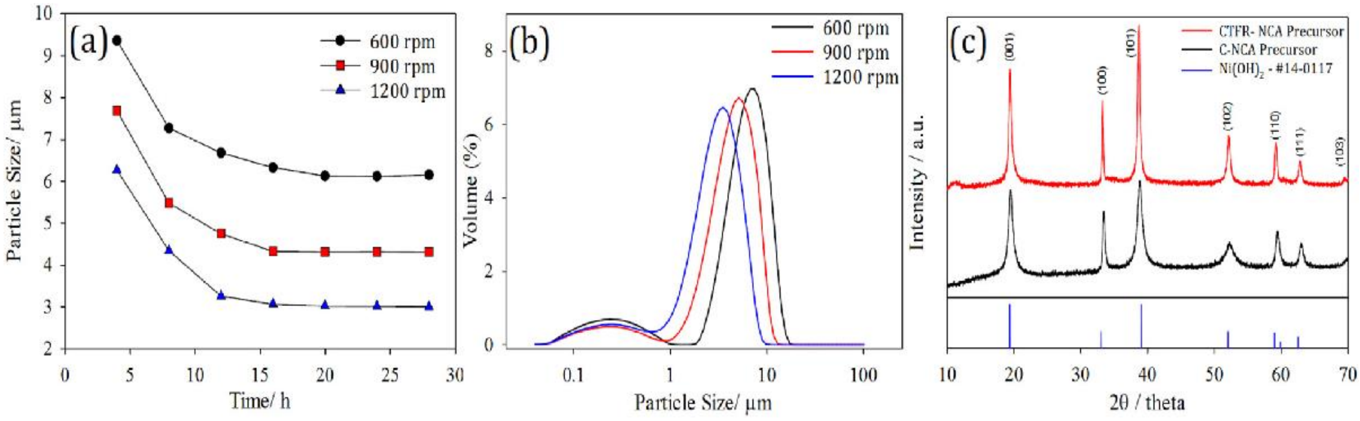
Better understanding of the effect of Co and Mn is expected to inform future NMC battery compositions for superior performance, sustainability, and safety.

Taylor Vortex Reactor for EV Battery Materials

3. Uniform and Stable NCA Hydroxide for Cathode Material

The Taylor Vortex Reactor’s uniform and constant mixing forces were used by M. Seenivasan et al. (2020) to prepare spherical and uniform $Ni_{0.80}Co_{0.15}Al_{0.05}(OH)_2$ particles with the following characteristics:

- Elemental mapping (EDX): Ni, Co, Al uniformly present in near-stoichiometric ratio of 78.5%, 16.1%, and 5.4%, respectively.
- Crystal structure (XRD): Diffraction peaks indexed to hexagonal phase of β-$Ni(OH)_2$. Lack of peaks for impurity phases indicates a pure phase product.
    - Rietveld refinement analysis further shows suggests that Taylor-made NCA hydroxide had highly ordered crystalline layers.
    - R-factor intensity ratios >1.2 indicate low cation mixing, small irreversible cation loss, and therefore, good electrochemical performance.
    - Intensity ratios of Taylor-made NCA hydroxide materials ranged from 1.810 to 1.639, indicating greater than recommended value and previously reported values NCA cation-mixing.

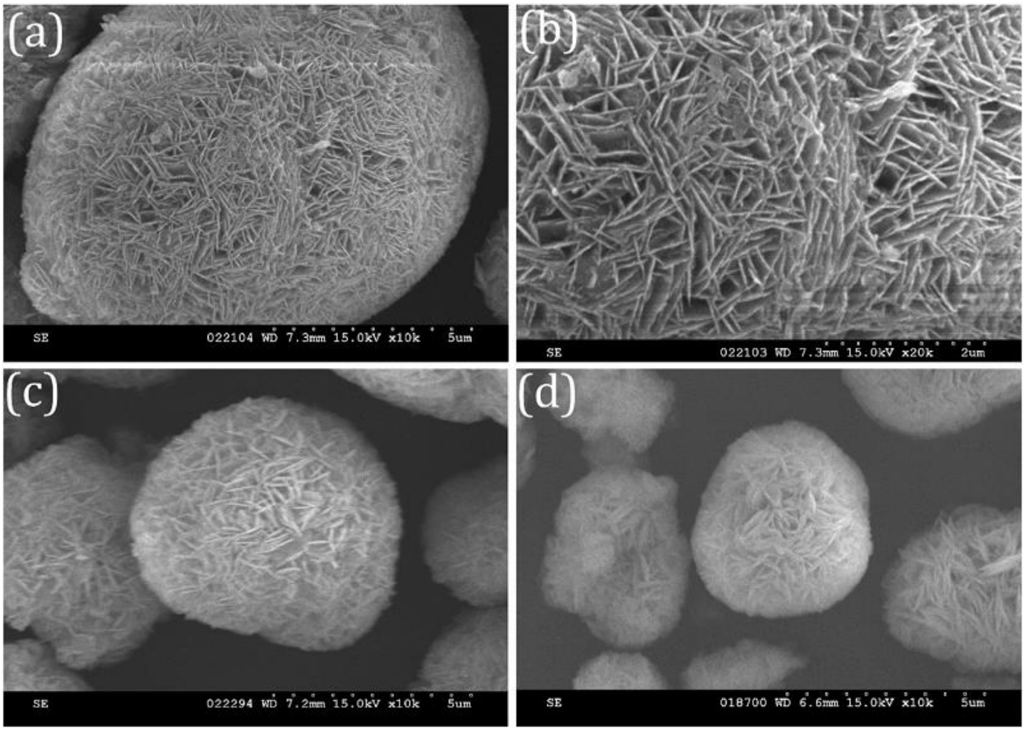


(a) Typical steady state flow behavior of NCA hydroxides in Taylor Crystallizer.
(b) Particle size distribution of NCA hydroxides prepared in Taylor Crystallizer at various speeds.
(c) Powder XRD patterns of Taylor-made NCA materials vs. commercial NCA material.

Taylor Vortex Reactor for EV Battery Materials

3. Uniform and Stable NCA Hydroxide for Cathode Material

- Morphology and microstructure (SEM, HR-TEM): spherical particles with average sizes of 5-6 µm. Polyhedral primary particles of sizes varying from 200 to 1 µm.
  - As smaller primary particle sizes indicate shorter diffusion pathways for Li^+ migration, they are a critical consideration for de/intercalation.

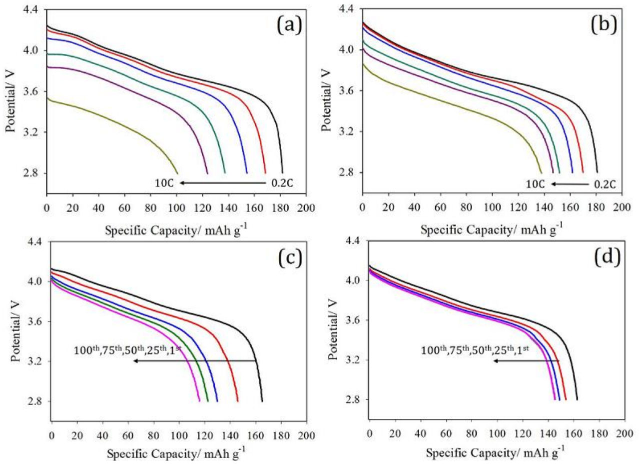


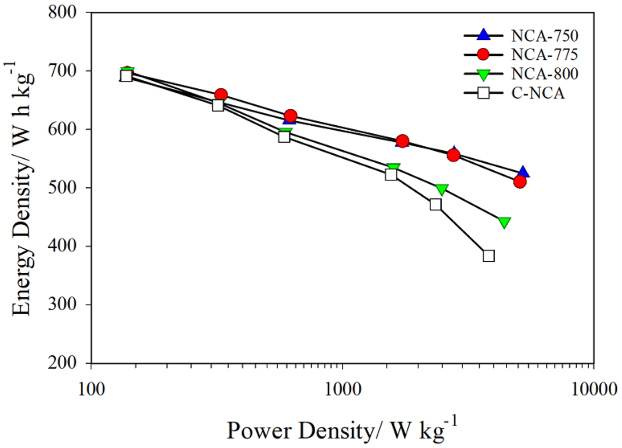
(a) SEM images of NCA Hydroxide materials prepared by Couette-Taylor Crystallizer at (a, b) 600 rpm, (c) 900 rpm, and (d) 1200 rpm.

Taylor Vortex Reactor for EV Battery Materials

3. Uniform and Stable NCA Hydroxide for Cathode Material

- Chemical composition (ICP-OES): experimental confirmation of stoichiometric compositions
- Electrochemical performance via preparation of CR2032 coin-type half-cells: rate capabilities in discharge range of 0.2-10C at 25°C with fixed charge rate at 0.2C
    - NCA-750 exhibited superior reversible capacity of 138.1 mAh/g at 10C. NCA-775 and NCA-800 exhibited capacities of 134.4 and 116.6 mAh/g, respectively.
    - All the Taylor-made samples demonstrated higher reversible capacities than cathodes prepared with commercially available NCA.
- Cycle Life over 100 cycles:
    - NCA-750 had initial capacity of 162.7 mAh/g and a 100th capacity of 145.1 mAh/g, for a retention of 87.4%. In comparison, the commercial NCA sample had a capacity retention of only 70.0%.





Left: Discharge voltage profiles of commercial NCA vs. Taylor-made NCA-750 sample

Right: Ragone plots for practical applicability of commercial NCA vs. Taylor-made NCA samples

4. Solid State Lithium Battery

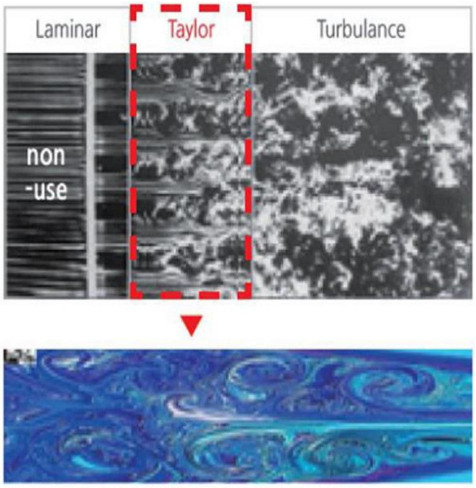
4.1 Ga-Doped LLZO Material

Researchers from KITECH (2017) compared the synthesis of Ga-doped LLZO for solid state batteries in a Taylor-Couette Reactor vs. a batch tank reactor.

While LLZO ($Li_7La_3Zr_2O_{12}$) is a metal oxide with excellent ionic conductivity and good electrochemical windows, it is difficult to prepare cubic-phase pellets with fine sintering properties.

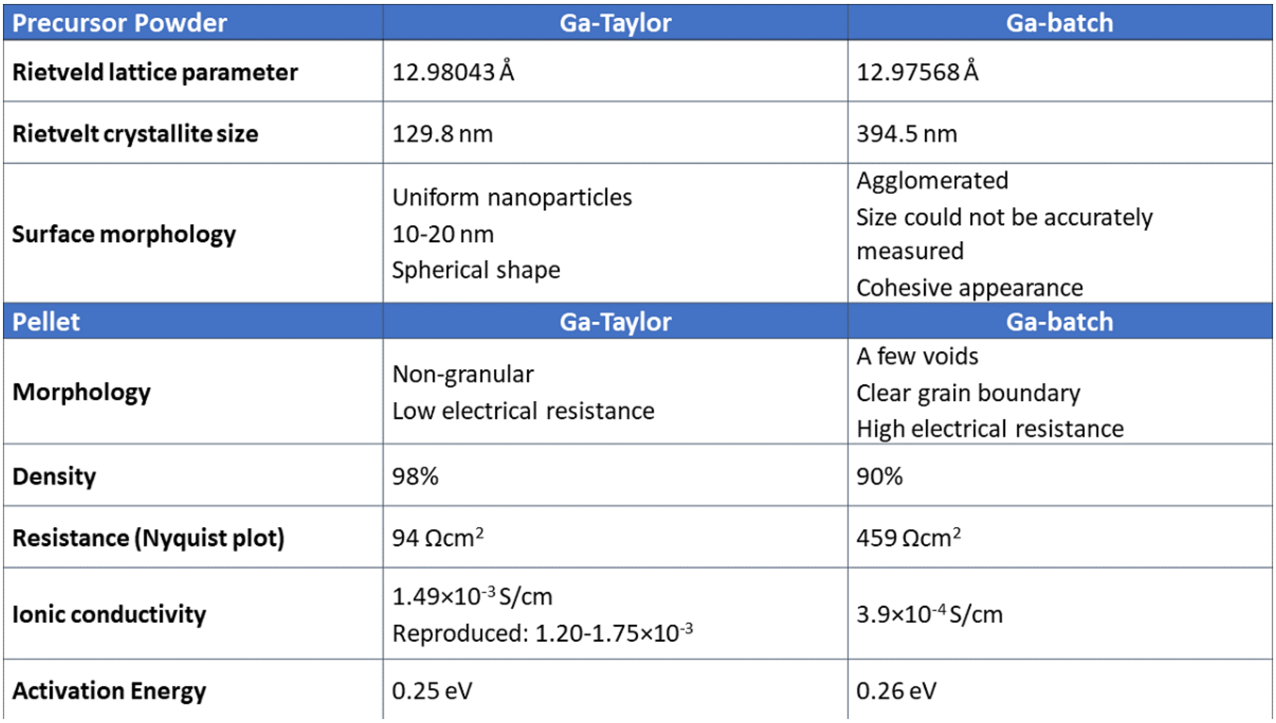
LLZO is particularly sensitive to process conditions, yet co-precipitation was thought to be a promising method for mass production. At the time of the publication, the highest ionic conductivity of LLZO prepared via conventional means is approximately 3-8 × 10^{-4} S/cm.

Yang et al. (2017) experimented with a new method of LLZO co-precipitation via the Taylor Flow Reactor to examine how the unique Taylor vortex fluid motion affects LLZO properties.



4.1 Ga-Doped LLZO Material

Yang et al. (2017) report that the toroidal vortex fluid motions created highly efficient radial mixing conditions, which led to reduced reaction time and increased yield. Simultaneously, the Taylor Vortex Reactor produced Ga-doped LLZO materials with lower crystallite size, and higher density and ionic conductivity, when compared to a batch reactor.

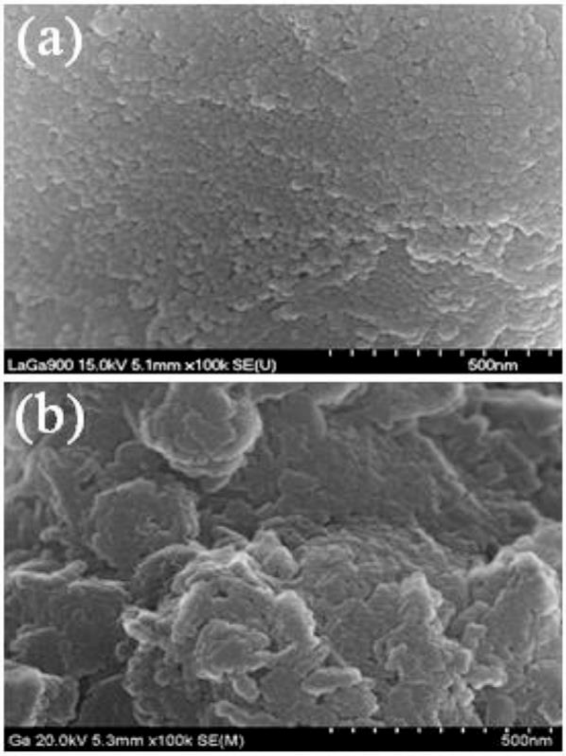


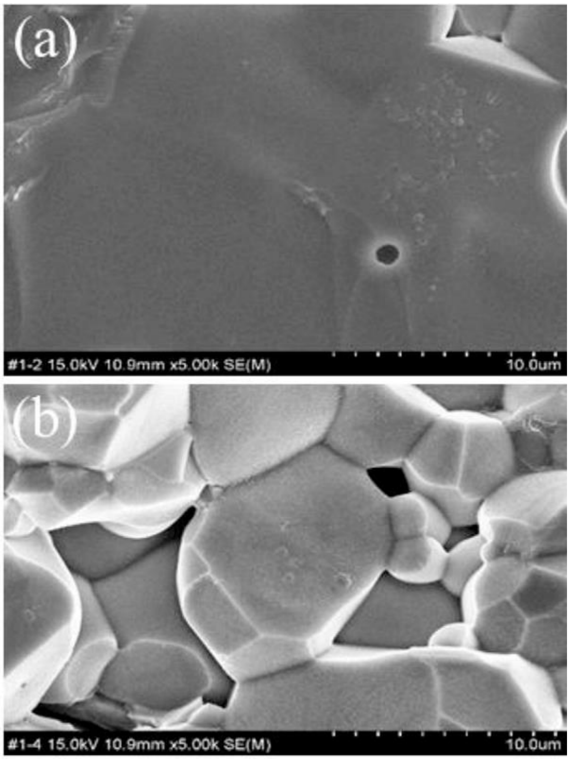
Precursor Powder	**Ga-Taylor**	**Ga-batch**
Rietveld lattice parameter	12.98043 Å	12.97568 Å
Rietvelt crystallite size	129.8 nm	394.5 nm
Surface morphology	Uniform nanoparticles 10-20 nm Spherical shape	Agglomerated Size could not be accurately measured Cohesive appearance
Pellet	**Ga-Taylor**	**Ga-batch**
Morphology	Non-granular Low electrical resistance	A few voids Clear grain boundary High electrical resistance
Density	98%	90%
Resistance (Nyquist plot)	94 Ωcm^2	459 Ωcm^2
Ionic conductivity	1.49×10^{-3} S/cm Reproduced: $1.20\text{-}1.75\times10^{-3}$	3.9×10^{-4} S/cm
Activation Energy	0.25 eV	0.26 eV

The toroidal vortex motion in the Taylor Crystallizer increased lattice parameter, which proportionately increased ionic conductivity while decreasing crystallite size. As seen in the values above, the total conductivity of the oxide-based LLZO pellet prepared in the Taylor Crystallizer approximates the high value of sulfide-based pellets.

4.1 Ga-Doped LLZO Material

In conclusion, Yang et al. (2017) demonstrated that the Taylor-Couette Crystallizer makes it possible to efficiently produce high-performing, spherical nanomaterials for solid state batteries.





Left: SEM Image of Ga-doped LLZO precursor synthesized in (a) Taylor Crystallizer and (b) Batch Reactor

Right: SEM Image of Ga-doped LLZO pellet synthesized in (a) Taylor Crystallizer and (b) Batch Reactor

4.2 Composite NCM-LZAOH Solid State Battery

In 2020, Heo et al. compared NCM80 and composite NCM80-LZAOH cathode materials for all-solid-state battery performance. Both the NCM80 and the composite NCM80-LZAOH material were synthesized in our 1L Taylor Vortex Reactor via co-precipitation.

The reactor was configured with a pH sensor that connects to the NaOH pump. The flow rate of the NaOH pump automatically adjusted throughout the reaction to maintain a reaction pH of 11 for 4 hours.

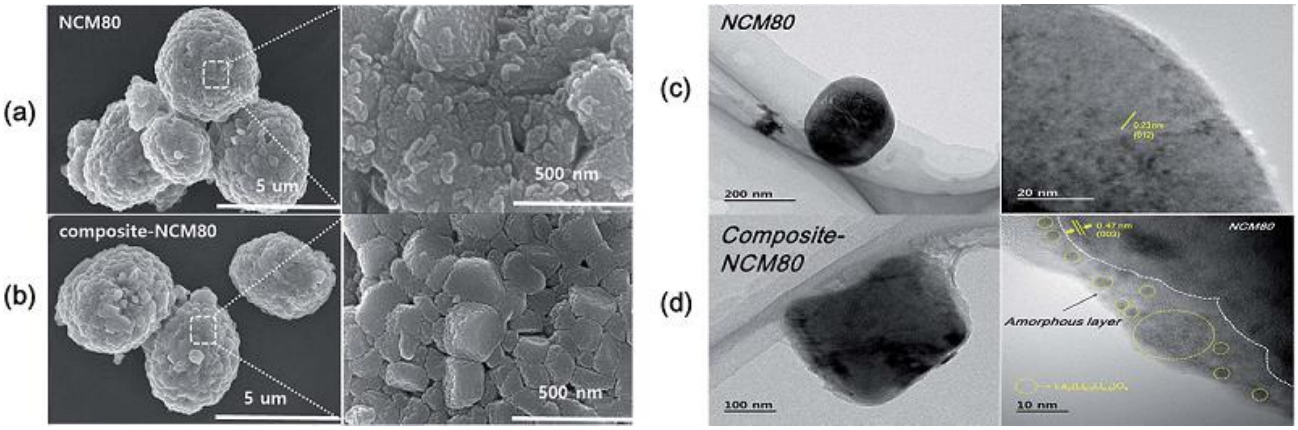
Results of NCM vs. Composite NCM-LZAOH Synthesis

FE-SEM images of both NCM80 and composite NCM-LZAOH materials display spherical particles with similar particle sizes of <5 µm. FE-TEM images further show that the primary particles were 100 to 200 nm in size.

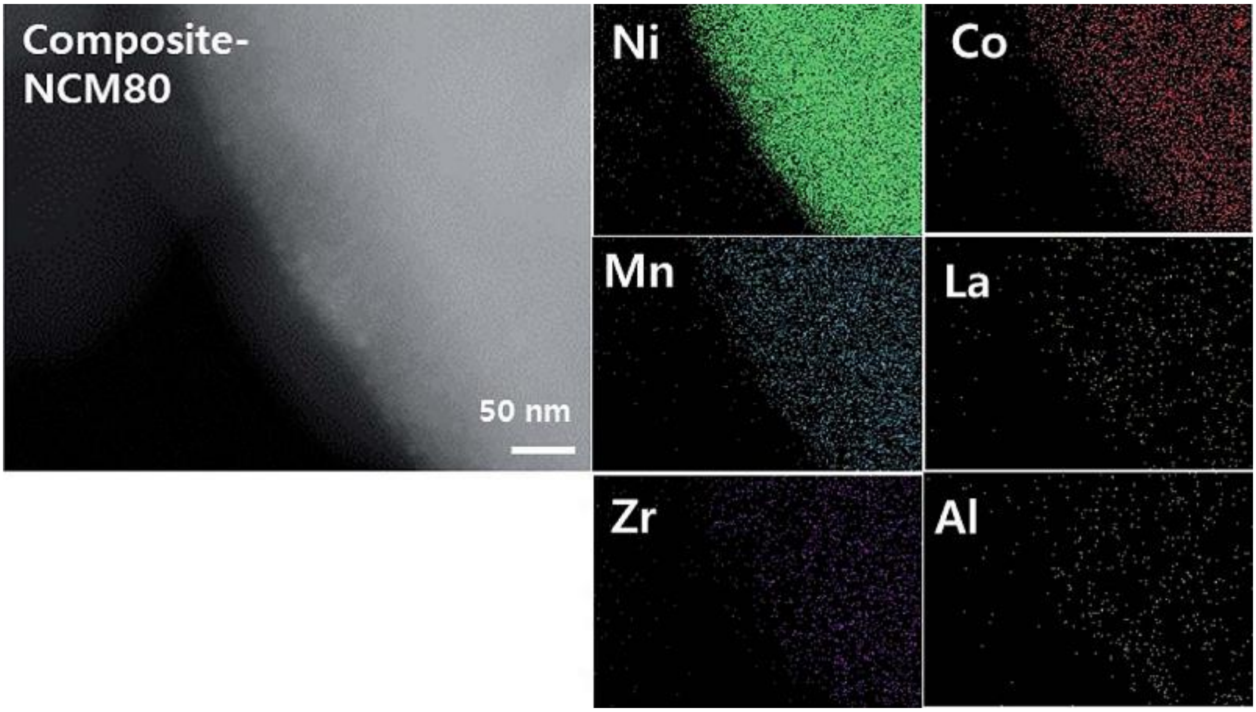
Additionally, TEM images show that the composite NCM-LZAOH material had a uniform coating layer roughly 10 nm thick, composed of nanomaterials 1-2 nm big, which was not present on NCM80 particles. EDS mapping images also demonstrated that the composite elements were uniformly distributed on the material surface, which indicates uniform coating of LLZAO material.

Images on next page.

4.2 Composite NCM-LZAOH Solid State Battery



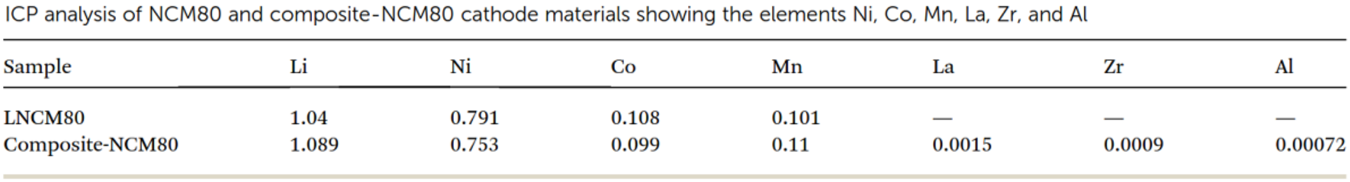
SEM images of (a) NCM80 and (b) composite NCM-LZAOH, and TEM images of (c) NCM80 and (b) composite NCM-LZAOH battery materials synthesized in Taylor Crystallizer



EDS mapping images display uniform distribution of Ni, Co, Mn, La, Zr, Al on coating of composite NCM-LZAOH material synthesized in Taylor-Couette Reactor

4.2 Composite NCM-LZAOH Solid State Battery

Furthermore, ICP analysis of both NCM80 and composite NCM-LZAOH reveal that the experimental atomic contents closely approximate the theoretical values. The ICP results indicates that the elements were uniformly distributed during co-precipitation in the Taylor Crystallizer.



ICP analysis of NCM80 and composite-NCM80 cathode materials showing the elements Ni, Co, Mn, La, Zr, and Al

Sample	Li	Ni	Co	Mn	La	Zr	Al
LNCM80	1.04	0.791	0.108	0.101	—	—	—
Composite-NCM80	1.089	0.753	0.099	0.11	0.0015	0.0009	0.00072

ICP analysis of NCM and Composite NCM-LZAOH material synthesized in Taylor Crystallizer reveal that experimental composition matches theoretical composition

Overall, the experiment found that the composite NCM-LZAOH cathode material presented higher initial discharge, initial coulombic efficiency, and discharge retention after 50 cycles when compared to the NCM80 material.

The study by Heo et al. (2020) is valuable in demonstrating the use of the Taylor-Couette Crystallizer to synthesize various particles with spherical morphology, uniform composition, and high electrochemical performance for next-generation solid state batteries.

Taylor Vortex Reactor for EV Battery Materials

5. Na-ion Cathode Material *via* Taylor-Couette Reactor

Finally, the Taylor-Couette Crystallizer’s ease of operation for manufacturing of new battery technology is well demonstrated by the 2017 study by Jo et al.

Prompted by concerns about high cost, low lithium availability and poor safety of Li-ion batteries, the researchers manufactured various Na-ion battery cathode materials. Layered Na-ion materials show improved conductivity and capacity with carbon coating and transition metal substitution.

By adjusting the reaction temperature of our Taylor-Couette Reactor, Jo et al. (2017) manufactured NaxMn[Fe(CN)6]•zH2O with varying crystal structures. They then evaluated the effects of varying Na and interstitial water content on the crystal structure and electrochemical properties of the Na-ion battery material.

Taylor Crystallizer Temperature Control Function

Na-ion battery materials were produced at a reaction temperature of 25°C and 60°C.

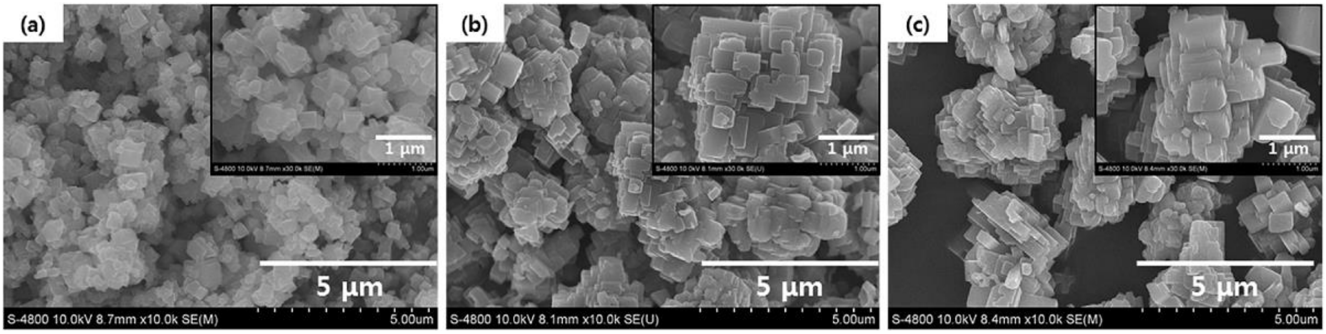
Our Taylor-Couette Crystallizer allows for adjustable reaction temperatures of up to 90°C. Our high-temperature model extends this control to up to 300°C.

The temperature of the reaction solution is control with our temperature regulating jackets that wrap around the entire exterior of the reaction tank. The high surface area of the cylindrical design of our Taylor Vortex Reactor allows for greater control and adjustability of the reaction temperature.

5. Na-ion Cathode Material via Taylor-Couette Reactor

Experimental Results of Na-ion Materials from Taylor-Couette Reactor

The temperature control function of the Taylor Vortex Reactor allowed the Jo et al. (2017) to examine the different crystal structures produced at varying temperatures and drying processes. In conclusion, they found that agglomeration of Na-ion cathode material was preferred at high reaction temperatures.



SEM images of Na-ion battery materials synthesized in the Taylor Crystallizer at various temperatures. (a) 25°C and air-dried, (b) 60°C and air-dried, (c) 60°C and vacuum-dried

Cathodes were prepared with a coating of the Na-ion material, then tested for electrochemical performance and stability. Overall, the cubic crystals synthesized at 25°C and air-dried displayed the highest capacity retention of 90.66% after 50 cycles.

The rhombohedral crystals, which were synthesized at 60°C and air-dried, had a capacity retention of 88.03% after 50 cycles. Moreover, the rhombohedral-structured crystals had a high initial reversible capacity of 150.1 mAh/g with a coulombic efficiency of ≥99.7%.

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