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Ultrafast charging of energy-dense lithium-ion batteries for urban air mobility

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ABSTRACT

Urban air mobility (UAM) demands batteries with high energy density, long cycle life, and fast rechargeability. Here, we demonstrate an energy-dense lithium-ion battery (LiB) with ultralong cycle life under ultrafast charging. By using the asymmetric temperature modulation (ATM) method, i.e., charging at an elevated temperature and discharging around the ambient temperature, it is experimentally shown that the 209 Wh/kg LiB is charged to 88% state of charge (SOC) in ~5 min under UAM cycling while retaining 97.7% capacity after 1,000 cycles. Moreover, an experimentally validated electrochemical-thermal (ECT) model is developed to elucidate the fast charging process and the degradation mode of UAM batteries, quantitatively capturing lithium plating during fast charging. We find that the LiBs for UAM applications are most prone to lithium plating due to their higher initial SOC required as the reserve for safety; nevertheless, the ATM method is effective in minimizing or preventing lithium plating in the high SOC range of 30-90%. In addition to slowing down capacity fade, the ATM method also raises the usable capacity by 10%, which boosts the battery energy density and ensures the battery to perform full UAM cycles even at the end of life.

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1. Introduction

Modern cities are afflicted with congestions and air pollution. Electric aircraft capable of taking off and landing freely in urban areas could be an ideal solution. Since 2017, NASA has been working together with industries to realize urban air mobility (UAM): a safe and efficient system for air passengers and cargo transportation within an urban area [1]. Till now, guite a few companies have shown and tested their prototypes of air taxis, and the majority of them are vertical takeoff and landing (VTOL) aircraft powered with electricity [2-5]. The electric powertrain is preferred because it is quiet, clean, and highly efficient [6,7]. Consequently, as the power source of the entire system, the performance of the battery becomes a critical factor in determining the market feasibility of electric VTOLs (eVTOLs).

Due to remarkable energy, capacity, and cost, lithium-ion batteries (LiBs) have become the most widely used electrochemical energy source today [8], powering up millions of battery electric vehicles (BEVs) on-road [9], which makes it a strong candidate for

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UAM applications as well. However, comparing with BEVs, eVTOLs have more stringent demands on specific energy, specific power, and thermal management of the battery pack [3]. State of the art BEV battery has reached a specific energy c.a. 250 Wh/kg, which could turn into 330 miles on a single charge [10]; while for eVTOLs, this energy density is still less satisfactory [6,7]. For eVTOLs with a 200-mile cruising range, the required specific energy goes up to 300 Wh/kg [3] at the pack level or ~400 Wh/kg at the cell level [7]. On the other hand, during takeoff and landing, eVTOLs require batteries to provide a sustained discharge rate above 3 C [3], indicating that one may not raise the internal resistance to increase the specific energy by using thick electrodes with low porosity and high mass loading [11]. Additionally, due to higher power consumption in UAM applications, heat generation of batteries in eVTOLs is predicted to be an order of magnitude larger than that in BEVs [3]. As such, besides pursuing higher specific energy for a single cell, developing a novel thermal management strategy is equally crucial to building lightweight battery packs for eVTOLs. Since the present battery technologies cannot meet all these requirements simultaneously, there has been scarce research on UAM batteries, and most of the relevant literature only involved theoretical calculations with assumed energy density [3,6,7,12–14]. Theoretically, the specific energy of LiBs could reach 400 Wh/kg









[8,11], but it will take years before such batteries become commercially available. The lack of energy density leads to a heavy battery pack on the aircraft and compromises the cruising range. Fredericks et al. [3] calculated the energy consumption of VTOL flights, suggesting that a 150 Wh/kg battery pack could fly a 50-mile eVTOL with four passengers under ideal conditions.

In this work, we propose an alternative to breaking this dilemma using an EV-type LiB with moderate energy density (209 Wh/kg) and compensating the short cruising range with ultrafast charging (5 min) strategy. We use a back-of-the-envelope calculation to illustrate the significance of fast charging for UAM applications and explain why fast charging can be used to make up for the lack of energy density. The concept of UAM, also known as on-demand urban air transport [6,7], requires high utilization of aircraft for profitability. The usage rate for a single unit could reach 3,000-5,000 h per year [6]. Rush hours in metropolises like New York City and Los Angeles usually last for 9 h each workday [15,16]. Assume an eVTOL fleet is fully loaded during the period: performing a 50-mile trip within 30 min and swapping passengers within 5 min repeatedly. If the battery needs a recharge and takes longer than 5 min, then the interval between two flights is determined by the battery's charging time. Fig. 1a depicts the timeline of eVTOLs during rush hours, given various combinations of cruising range and charge time. Long-range eVTOLs demonstrate a prominent advantage in efficiency when it takes an hour to recharge the battery. However, if we could accomplish the recharge within 5 min, the usage rate of the eVTOLs becomes independent of the cruising range, and at the same time, max out the unit efficiency. Fig. 1b plots the number of trips during rush hours vs. charging time. It is seen that the maximum usage rate of eVTOLs is



Fig. 1. The relationship between cruising range, fast charging capability, and usage rate for eVTOLs a) Timeline of eVTOLs with different combinations of cruising range and charge time during rush hours. b) The maximum trips per day during rush hours. The trip number is strongly related to the cruising range under low charge rates; if the recharge of the battery could be accomplished within 5 min, the usage rate of eVTOLs is not affected by the cruising range.

determined by the charging time of the battery. For batteries capable of fast charging within 5 min, they could support fully loaded eVTOLs to complete 15 trips in 9 h, and the usage rate during rush hours is around 2,000 h per year (0.5 h/trip×15 trips/work-day×260 workdays/year). Adding up with the flights in the rest of the time, the total usage rate in a year could reach 3,000 h or more. It is worth noting that the high usage rate of eVTOLs also puts forward higher requirements on the cycle life of the batteries; thus, minimizing the damage caused by fast charging is critical to succeeding in UAM applications.

During fast charging, the most deleterious aging mechanism is lithium plating [17,18]. If an applied charge rate exceeds the rate capability of lithium insertion and ion transport, lithium ions will deposit on the surface of negative electrodes in metallic form, which is the so-called lithium plating [19]. Since the deposited lithium metal reacts with electrolyte and leads to loss of recyclable lithium, the cell capacity would fade rapidly once lithium plating occurs [17,20]. Another adverse effect of lithium plating is dendrite growth [17]; the dendrite could penetrate the separator and cause an internal short-circuit, which is a hazardous condition for the battery [21,22]. Previous studies provided several ways to mitigate lithium plating at high-rate charging, which can be divided into two categories. The first one is to optimize charging algorithms [23]. For example, Sobana et al. [24] embedded a lithium titanate reference electrode into a LiB and kept graphite electrode potential to be 20 mV vs. the reference electrode when charging the cell. The charging current reached the maximum at the beginning and decreased at higher SOC. The current profile during this anode potential control reveals the full capability of the charge rate through the charging process, which decreases with SOC. Consequently, charging strategies with declining charge rates, such as multistage constant current (MCC) charging [24] and boostcharging [25], could somewhat maneuver charging without Li plating. However, charge algorithms are unable to break away the fundamental limitations posed by kinetics and transport properties of a LiB. The second approach is to improve the electrochemical properties of the LiB; some studies introduced new chemistries with improved kinetics/transport properties [26–28] to enhance the fast charging performance. Alternatively, rapid preheating the LiBs to elevated temperatures prior to fast charging brings an allaround improvement in kinetics and ion-transport processes without modifying materials of LiBs [29,30].

Besides lithium plating, mechanical degradation is another potential degradation mechanism for aging during fast charging [23]. LiBs operating at high rates experience severer nonuniformity in lithium concentration within active material particles [30,31]. Consequently, fast charging could build up higher strain inside the particles and generate more mechanical loss than moderate charge rates [31,32]. Improving the solid ion diffusivity through elevated temperatures would be an effective way to relieve mechanical degradation, which is also crucial to avoid lithium plating on particle surfaces.

Recently, we developed the asymmetric temperature modulation (ATM) method to enable extreme fast charging (XFC) of a BEV battery [33]. The technique involves preheating the cell to ~60 °C prior to fast charging so as to enhance kinetic and mass transport properties such that lithium plating could be minimized or prevented. Except for the 10-min fast charging period, the cell is otherwise maintained at ambient temperature, and consequently, the high-temperature degradation is limited to short time exposure [33]. We apply the same approach in this work to perform 5-min charging in a UAM cycle. In what follows, we first propose a fast charging UAM cycling protocol representative of eVTOL applications. Subsequently, we experimentally illustrate how a 209 Wh/kg LiB achieves 5-min charging with long cycle life for UAM applications. Finally, a validated electrochemical-thermal model is developed and solved in GT-Autolion software [34] to predict UAM battery behaviors under various situations and provide important insights.

2. UAM cycling protocol

We begin with the definition of a cycling protocol representative of UAM applications. Its charging part follows a constant current constant voltage (CCCV) profile where the charge rate is 6 C with a cutoff voltage of 4.15 V. Similar charging profiles have been used in generic XFC tests; however, the initial SOC and the end-of-charge conditions differ for UAM application. XFC cells usually cycle between 0% to 80% SOC [33,35]. In comparison, UAM cells require a reserve in energy and a minimum SOC around 30% [3,7,12]. The maximum SOC should be as high as possible to increase usable energy [7]. There are two criteria to determine the termination of the charging step: 1) charging time reaches 5 min, 2) charge current drops below 3 C. When both conditions are met, we will terminate the charging, as shown in Fig. 2a.

The UAM discharge protocol consists of a typical eVTOL flight of which the cruising range and altitude are shown in Fig. 2b, covering a 50-mile mission range at 1,000 feet above ground level (AGL) [36], and a 5-mile divert at 500 feet AGL after balked landing [37]. Based on the operating condition of eVTOLs, we divide the mission profile into nine segments, where A-E corresponds to the 50-mile mission range, and b-e corresponds to the 5-mile divert. Every one of them corresponds to a constant power discharge step for the battery pack. The magnitude of discharge power is calculated with a similar approach presented by Fredericks et al. [3] and Brown et al. [38]; the parameters used in the calculation are shown in Table 1.

Table 1

Parameters to calculate operating condition.	
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Parameter	Description	Value
W/A [N/m ²]	disk loading (ratio of weight to total rotor area)	343
ρ [kg/m³]	air density	1.225
η _{mech}	electromechanical efficiency	0.95
η _{prop}	propeller efficiency	0.9
f	fuselage down-wash correction	1.03
FoM	figure of merit	0.7
(L/D) _{max}	lift-to-drag ratio during cruise	14
(L/D) _{climb}	lift-to-drag ratio during climb	12
(L/D) _{descend}	lift-to-drag ratio during descend	12

Vertical climb/hover/descend power:

$$\frac{P}{W} = \left[\frac{f}{FOM}\sqrt{\frac{f(W/A)}{2\rho}} + \frac{V_{v}}{2}\right] / \eta_{mech}$$
(1)

Fixed-wing climb/cruise/descend power:

$$\frac{P}{W} = \left[V_{\rm v} + \frac{V}{L/D} \right] / \left(\eta_{mech} \eta_{prop} \right) \tag{2}$$

Cruise speed and minimum power speed:

$$V_{loiter} = [1/3]^{1/4} V_{cruise}$$
 (3)

The cruise speed is set to be 150 mph [37,38]. The horizontal speed during climb and descend is set to be the loiter speed of 114 mph, according to Eq. (3). The rate of climb and the rate of descent are both set to be 500 fpm [37]. The vertical speed during vertical takeoff and landing should be less than 500 fpm [37], which



Fig. 2. UAM cycling protocol. a) Charge profile: 6 C, 4.15 V CCCV charge; the end of charge is triggered when charge time \geq 5 min and charge current \leq 3 C. b) Flight profile of eVTOLs, including a 50-mile mission at 1,000 feet AGL and a 5-mile divert at 500 feet AGL. c) Discharge profile developed based on the flight profile in b. d) Payload vs. pack-level energy density of the battery.

contributes to less than 6% of the hover power and is assumed to be negligible to simplify the protocol.

I. Segment A: Vertical takeoff & hover. V = 0, $V_v = 0$. Step last for 30 s [36].

According to Eq. (1), P/W = 18.6 W/N.

II. Segment B: Fixed-wing climb. V = 114 mph, $V_{\nu} = 500$ fpm. Step takes 2 min to reach the cruise altitude of 1,000 ft [36].

According to Eq. (2), P/W = 7.94 W/N.

III. Segment C: Fixed-wing cruise. V = 150 mph, $V_v = 0$. Step takes 17 min to reach the 50-mile mission range.

According to Eq. (2), P/W = 5.60 W/N.

IV. Segment D: Fixed-wing descend. V = 114 mph, $V_v = -500$ fpm. Step takes 2 min to approach zero altitude.

According to Eq. (2), P/W = 2 W/N.

V. Segment E: Hover and vertical landing. V = 0, $V_v = 0$. Step last for 30 s [36].

According to Eq. (1), P/W = 18.6 W/N.

VI. Segment b: Fixed-wing climb (divert). V = 114 mph, $V_v = 500$ fpm. Step takes 1 min to reach the cruise altitude of 500 ft [37].

According to Eq. (2), P/W = 7.94 W/N.

VII. Segment c: Fixed-wing cruise (divert). V = 150 mph, $V_v = 0$. Step takes 30 s to reach the 5-mile divert distance.

According to Eq. (2), P/W = 5.60 W/N.

VIII. Segment d: Fixed-wing descend (divert). V = 114 mph, $V_v = -500$ fpm. Step takes 1 min to approach zero altitude.

According to Eq. (2), P/W = 2 W/N.

IX. Segment d: Hover and vertical landing (divert). V = 0, $V_v = 0$. Step last for 1 min.

According to Eq. (1), P/W = 18.6 W/N.

We can convert the required power for operation (*P*/*W*) to the discharge power of the battery (*P*/*E*_{batt}) with a constant conversion factor, *E*_{batt}/*W*, which represents distributed battery energy (C/3 rated energy for a fresh LiB) per unit weight of the aircraft. We set the value of *E*_{batt}/*W* to be 6.2 Wh/N so that the maximum discharge power for a fresh battery is 3 W/Wh. We can attain the discharge power for every step with the conversion factor; if we integrate the discharge power with time, we could find that the discharge energy takes up about 45% of the rated battery energy. The choice of *E*_{batt}/*W* is case-specific; a larger *E*_{batt}/*W* value helps to decrease the maximum discharge rate and the portion of energy consumption but undermines the payload and vice versa.

Fig. 2c shows the complete discharge profile used in this work. The discharge power is normalized with the C/3 discharge energy of a fresh cell and is kept unchanged throughout the lifetime of LiBs. This power protocol is similar in form to the federal urban driving schedule (FUDS) for electric cars [39,40], after removing

regenerative braking pulses in FUDS. We summarize the detailed specifications for discharge in Table 2.

The pack size and the energy density of a battery come into play when calculating the payload. First, we can get the gross takeoff mass (GTOM) of the aircraft with Eq. (4). Where E_{batt} denotes the energy of a battery pack and equals 140 kWh [6] (rated at C/3 discharge); E_{batt}/W is a conversion factor between discharge power and workload for the aircraft, which equals 6.2 Wh/N according to the supplementary information; the standard gravity, *g*, equals to 9.8 m/s². Substituting the values into the formula, we can get the GTOM is about 2,300 kg.

$$\text{GTOM} = \frac{E_{batt}}{(E_{batt}/W)g} \tag{4}$$

Based on the GTOM of the aircraft, we can calculate the payload with Eq. (5). The empty weight ratio, W_e/W , is a design parameter for aircraft and is set to be 0.55 [3]. The payload can then be expressed as a function of the pack-level energy density, e_{batt} , as shown in Eq. (5) and Fig. 2d.

$$m_{pay} = (1 - W_e / W) \text{GTOM} - E_{batt} / e_{batt}$$
(5)

An important issue to consider is the cell to pack (CTP) integration efficiency. To date, the gravimetric CTP ratio (cell specific energy over pack specific energy) of commercial EVs is ~75% (e.g., BMW i3 with prismatic cells [41]). Thus, a battery pack with this work's cells (209 Wh/kg rated at C/3) has a specific energy of ~150 Wh/kg, which corresponds to ~100 kg payload or one passenger (Fig. 2d). A further increase of payload requires advancement not only in cell specific energy but also in CTP efficiency. Very recently, battery suppliers like CATL and BYD have developed socalled CTP technology that directly assembles cells into a pack without using modules and thus increases the gravimetric CTP ratio to as high as 85% [42]. Furthermore, we note that the method of operating batteries at elevated temperatures can reduce battery cooling needs by >12x due to enlarged temperature difference between the cell and ambient and reduced heat generation rate associated with the lower cell internal resistance, as analyzed detailedly in our recent works [33,43]. As such, the battery thermal management system (BTMS), which takes ~5% of pack weight [44], can be greatly simplified or even eliminated. Combining the CTP technology with a simplified BTMS, we project a gravimetric CTP ratio of 90%, resulting in a pack-level energy density of ~190 Wh/kg with this work's cells and, accordingly, a payload of ~300 kg (Fig. 2d).

3. Experimental

Pouch cells with thick electrodes are fabricated to conduct an experimental study. The negative electrode made of graphite has an areal capacity of 2.94 mAh/cm^2 . The positive electrode consists of LiFePO₄ (LFP) coated LiNi_{0.5}Mn_{0.3}Co_{0.2}O₂ (NMC532) with an areal capacity of 2.55 mAh/cm^2 . We fabricate pouch cells in 5.2 Ah and 35 Ah formats with the same kind of electrodes. The 5.2 Ah-format cells are selected to perform UAM cycling tests; the 35 Ah-format cells are used to demonstrate the cell-level specific energy as 209 Wh/kg, which we use to denote the cell-level energy density of the employed electrodes. Detailed specifications of the cells are listed in Table 3.

Fig. 3a sketches the experimental setup. The positive and negative terminals are connected to an electronic load to perform cycling tests. The third terminal, called the ACT terminal, is connected to the positive terminal with a relay to realize preheating prior to charging. The cell has a heating element embedded in the electrode stack during fabrication [45-47] with one side connected

Table 2

Mission profile and discharge power for the electric VTOLs.

Segment	Time, [s]	Horizontal speed, [mph]	Vertical speed, [fpm]	Power, [W/N]	Power, [W/Wh]	Operating condition of the electric VTOLs
Α	30	0	0	18.6	-3	Takeoff & Hover
В	120	114	500	7.94	-1.28	Fixed-wing climb
С	1020	150	0	5.60	-0.9	Fixed-wing cruise
D	120	114	-500	2.00	-0.32	Fixed-wing descend
E	30	0	0	18.6	-3	Hover & Landing
b	60	114	500	7.94	-1.28	Fixed-wing climb
с	30	150	0	5.60	-0.9	Fixed-wing cruise
d	60	114	-500	2.00	-0.32	Fixed-wing descend
e	60	0	0	18.6	-3	2nd Hover & Landing

Table 3

Cell design information of 5.2Ah cell and 35Ah cell.

Name	Sub-name	5.2 Ah cell	35 Ah cell		
Anode	Active material	Artificial Graphite			
	BET area	1.5 m ² /g			
	Mass Loading	8.39 mg/cm ²			
	Areal capacity	2.94 mAh/cm ²			
	Cu foil thickness	8 µm			
	Dimension	123×72 mm	196×154 mm		
	Number of layers	14	25		
Cathode	Active material	LiFePO ₄ -coated (10 wt%)			
		LiNi _{0.5} Mn _{0.3} Co _{0.2} O ₂ (90 wt%)			
	Mass Loading	15.92 mg/cm ²			
	Areal capacity	2.55 mAh/cm ²			
	Al foil thickness	15 µm			
	Dimension	120×69 mm	192×151 mm		
	Number of layers	13	24		
Electrolyte		1M LiPF ₆ dissolved in EC/EMC			
		(3:7 by wt.)+2wt	% VC		
Separator		Celgard-2325 microporous tri-			
		layer membrane			
Weight		114 g	619 g		
Capacity		5.2 Ah	35 Ah		
Specific Energy	C/3	164 Wh/kg	209 Wh/kg		
	1 C	155 Wh/kg	196 Wh/kg		

to the negative terminal and the other side connected to the ACT terminal; the added weight due to the heating element is 1.5% of the cell [45]. The working principle of this self-heating LiB is illustrated in Fig. 3b: if the relay is open, no current will flow through the heating element, and the cell behaves the same as a conventional LiB. When the relay is closed, a current will flow through the heating element due to the potential difference between positive and negative terminals; the current can either come from the charger or be discharged by the cell.

4. Electrochemical-thermal modeling

We build an electrochemical-thermal (ECT) coupled model to explore the behaviors of this UAM battery under various conditions. Governing equations of this ECT model, with Li diffusion in active material particles described by the so-called pseudo-2D model, have been widely introduced in our previous work [48–50] and solved with GT-Autolion, a commercial battery simulation package [34]. We adjust the input parameters to match the simulation results with the experimental discharge curves at various C-rates, as shown in Fig. S1. The governing equations [40,51–53] are given below, and the modeling parameters [54–63] are given in the supplementary information (Tables S1–S2 and Figs. S2–S3).

The reaction rate is related to the surface overpotential by the Butler-Volmer equation.

$$\vec{u} = i_0 \left[\exp\left(\frac{\alpha_a F}{RT} \eta\right) - \exp\left(-\frac{\alpha_c F}{RT} \eta\right) \right]$$
(6)

- *i*: reaction current density.
- *i*₀: exchange current density.
- α_{a} , α_{c} : (anodic/cathodic) apparent transfer coefficients.
- F: Faraday constant.
- *R*: universal gas constant.
- *T*: absolute temperature
- η : surface overpotential.

Surface overpotential.

$$\eta = \Phi_s - \Phi_e - U - iR_f \tag{7}$$



Fig. 3. Experimental setup for UAM cycling. a) Schematic of cell connections in UAM cycling experiments. b) An equivalent circuit for the test setup.



Fig. 4. Illustration of asymmetric temperature modulation (ATM) method for a UAM cycle. (a-d) Preheating to 60 °C with nickel foil by charge current. (e-h) 5-min charging at ~60 °C. (i-l) Temperature and voltage evolutions during UAM discharge.

- Φ_s , Φ_e : solid phase/electrolyte potential
- *U*: equilibrium potential vs. $E^{\circ}_{Li|Li+}$
- *R_f*: film resistance.

Volumetric current density can be obtained by multiplying a factor with the electrode current density.

$$j = a_s i, \ a_s = 3(1 - \varepsilon)/r_i \tag{8}$$

- *a*_s: specific surface area (SSA)
- ε: porosity
- *r_i*: particle radius

Effective parameters in a porous medium.

$$\Psi_s^{\text{eff}} = (1 - \varepsilon)^p \Psi_s, \Psi_e^{\text{eff}} = \varepsilon^p \Psi_e \tag{9}$$

- Ψ_{s} , Ψ_{e} : solid/electrolyte phase parameters
- P: Bruggeman exponent

Charge conservation in the solid phase.

$$\frac{dI_s}{dx} = \frac{d}{dx} \left(-\sigma^{eff} \frac{d\Phi_s}{dx} \right) = -j \tag{10}$$

- I_s : superficial current density in solid phase
- *σ*: electrical conductivity



Fig. 5. Cycling behaviors of ATM and baseline cells. a) Capacity retention vs. cycle number. The ATM cell has a capacity loss of 2.3% after 1,000 cycles, while the baseline cell only lasts for ~150 cycles. b) Minimum voltage during UAM discharge. The ATM cell has 170 mV voltage decay after 1,000 cycles, while the baseline cell cannot complete the discharge protocol after 155 cycles. The voltage decay originates from both capacity loss and internal resistance rise. c) Relaxation voltage of the cell during rest immediately after fast charging. A voltage plateau can be observed for the baseline cell between 30 and 90 s. d) Differential voltage during rest after fast charging shows a 90-s Li-stripping process, indicating that severe lithium plating has occurred in the baseline cell.



Fig. 6. Measured and predicted cell voltage and temperature evolutions for fresh cells. a, b) ATM cell with preheating before charging. c, d) baseline cell without preheating.



Fig. 7. Evolution of equilibrium potential and surface overpotentials at the negative-electrode separator interface (NSI). a) Fast charging a partially charged cell without preheating (baseline cell). Mass transport loss comes from ion transport in solid particles and electrolyte. Large surface overpotential for lithium insertion leads to lithium plating. b) Fast charging a partially charged cell with preheating (ATM cell). c) Fast charging a low SOC cell without preheating. d) Fast charging a low SOC cell with preheating. e, f) Evolution of cell temperatures.

Charge conservation in the electrolyte.

$$\frac{dI_e}{dx} = \frac{d}{dx} \left(-\kappa^{eff} \frac{d\Phi_e}{dx} - \kappa_D^{eff} \frac{d\ln c_e}{dx} \right) = j$$
(11)

- *I_e*: superficial current density in electrolyte
- *κ*: ionic conductivity
- *κ*_D: diffusional ionic conductivity
- *c*_e: electrolyte concentration

Diffusional ionic conductivity.

$$\kappa_D^{eff} = \frac{2RT\kappa^{eff}}{F} \left(t_+^0 - 1 \right) \left(1 + \frac{d\ln f_\pm}{d\ln c_e} \right) \tag{12}$$

• t_+ : transference number of Li-ion

• f_{\pm} : activity coefficient

Species conservation in the electrolyte.

$$\frac{d}{dt}(\varepsilon c_e) = \frac{d}{dx} \left(D_e^{eff} \frac{dc_e}{dx} \right) + \left(\frac{1 - t_+^0}{F} \right) j \tag{13}$$



Fig. 8. Voltage and current vs. SOC during UAM cycling. a) Evolution of cell voltage within balanced SOC windows. ATM method reduces the charging overpotential dramatically so that the cell could be charged to a higher SOC window. b) Evolution of charge/discharge current within balanced SOC windows.

• De: ionic diffusivity

Species conservation in the active material particles is given by Fick's law.

$$\frac{\partial c_s}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 D_s \frac{\partial c_s}{\partial r} \right) \tag{14}$$

• *D_s*: solid diffusivity

• *c*_s: ion concentration in solid phase

Heat generation is calculated by the equation below. The heat generation comes from four components, namely: heat generation at the reaction interface, heat generation from the solid electrodes, heat generation from the electrode, and reversible heat.

$$\dot{Q}_{gen} = A_E \int_0^L \left(\dot{q}_i + \dot{q}_s + \dot{q}_e + \dot{q}_{rev} \right) dx$$

$$\dot{q}_i = j(\Phi_s - \Phi_e - U)$$

$$\dot{q}_s = I_s \left(-\frac{d\Phi_s}{dx} \right) = \sigma_s^{eff} \left(\frac{d\Phi_s}{dx} \right)^2 \qquad (15)$$

$$\dot{q}_e = I_e \left(-\frac{d\Phi_e}{dx} \right) = \kappa^{eff} \left(\frac{d\Phi_e}{dx} \right)^2 + \kappa_D^{eff} \frac{d\ln c_e}{dx} \frac{d\Phi_e}{dx}$$

$$\dot{q}_{rev} = j \left(T \frac{dU}{dT} \right)$$

• *A_E*: electrode area

Arrhenius equation

$$\Psi(T) = \Psi_{ref} \exp\left(\frac{E_{act}}{R} \left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right)$$
(16)

• *E*_{act}: activation energy

5. Results and discussion

5.1. Fast charging of a UAM cell

A single UAM cycling experiment based on the ATM method is shown in Fig. 4 for a specific description. Prior to fast charging, the cell is preheated from room temperature to 60 °C using a 5 C charge current from the charger, taking about 50 s. The voltage drop through the relay and the heating element roughly equals the open-circuit voltage (OCV) of the cell. As shown in Fig. 4c, there is only a minor change of cell voltage when preheating begins, indicating that the discharge current from the cell is negligibly small. Once the cell temperature reaches 60 °C, the relay is open, and heating is stopped while cell charging commences, with the cell temperature maintained around 60 °C to minimize lithium plating.

The interplay between heat generation and dissipation determines the cell temperature evolution. Since heat generation, directly proportional to the much lower internal resistance at elevated temperature, is reduced, and additionally, the temperature difference between the cell surface and the environment is much enlarged, the need for cooling during fast charging is significantly reduced. The fast charging step lasts for 5 min, and the UAM discharge profile begins immediately after that. The magnitude of the discharge current is much smaller than the fast charging current; consequently, heat dissipation overwhelms the heat generation under the same thermal boundary conditions, leading to a dramatic and fast drop in cell temperature during discharge steps.

We put two identical cells under cycling according to the UAM protocol described earlier. The first cell adopts a preheating step prior to charging (labeled as 'ATM'), while the second cell is charged and discharged without preheating, which is marked as the baseline case. For every 50 or 100 cycles, we would pause the cycling tests to calibrate capacity retention with a reference performance test (RPT), where a C/3 CCCV protocol (4.15 V cutoff voltage and C/ 20 cutoff current) is used to fully charge the cell, and then the discharge capacity is measured with C/3 rate to 2.7 V. The capacity retention and the minimum discharge voltage are plotted vs. cvcle number, as displayed in Fig. 5a and b. The ATM method significantly improved the cycle life of the cell. The ATM cell only has a capacity loss of 2.3% after 1,000 UAM cycles. In contrast, the capacity retention of the baseline cell drops to 80% after about 150 cycles, and the minimum voltage becomes less than 2.7 V at the 155th cycle. Note that cycling of the UAM cell was shut down around 800 cycles due to COVID-19 and resumed after two months; during the long rest period, the lithium stoichiometry and electrolyte could equilibrate in the in-plane direction, which explained a little pickup in capacity retention in Fig. 5a.

Lithium plating is detected in the baseline cell through a voltage relaxation method [64,65]. We record the relaxation voltage of the battery during a 5-min rest period immediately after charging and plot its derivative with respect to time. Usually, we would expect the differential voltage to decrease monotonically with time, as the



Fig. 9. SOC window under UAM cycling. a) End of charge (EOC) SOC vs. initial SOC. The curve consists of two segments. The first segment represents the charging process terminated with the cutoff current of 3 C (and charging time > 5 min); the second segment represents the charging process terminated with 5 min charging time (and cutoff current < 3 C). b) End of discharge (EOD) capacity vs. initial SOC. The discharge voltage drops below 2.7 V when EOD SOC goes below 15%. c) Calculation of balanced SOC window for an ATM cell. d) Calculation of balanced SOC window for a baseline cell.

gradient of Li-ion concentration in the solid particles and electrolyte relax with time. However, if lithium plating occurs during charging, Li-stripping will dominant at the initial stage of rest and hold the negative electrode potential at a constant value. Consequently, the voltage relaxation of the graphite electrode will be postponed until the Li-stripping ends, which creates a peak on the differential voltage curve. As shown in Fig. 5c, a voltage plateau can be observed on the voltage curve of the baseline cell, and the peak labeled on Fig. 5d is evidence of severe lithium plating [64]. The result proves that the short cycle life of the baseline cell is caused by lithium plating. On the contrary, the ATM cell shows no sign of lithium plating, explaining why its 5-min charging cycle has a long cycle life.

5.2. Modeling results of lithium plating

With the experimentally validated ECT model, we simulate the LiB during a UAM cycle under different thermal conditions. As shown in Fig. 6, the numerical results are plotted along with the experimental results. Fig. 6a, b displays a UAM cycle with ATM method (insulated b.c. or $h = 10 \text{ W/m}^2\text{K}$). Fig. 6c, d shows the baseline case. The cell is charged at room temperature without preheating (under natural convection or $h = 20 \text{ W/m}^2\text{K}$). The predicted voltage and temperature profiles are in close agreement with the experimental results in both cases. It is worth mentioning that fast-charging is commonly thought of requiring much stronger cooling. A significant advantage of the ATM method is that charging at an elevated cell temperature of 60 °C can reduce the battery cooling need by >12× compared with fast charging at room temperature, as detailed in Ref. [33]. This is because of a reduced heat

generation at 60 °C due to the lower internal resistance, and of the enlarged temperature difference between the cell and ambient driving heat dissipation.

During charging, the surface overpotential of lithium deposition/stripping reaction on the graphite electrode can be derived from Eq. (7), as given below. Where *U* denotes the equilibrium potential of Li insertion into graphite particles, and $\eta_{LixC6|Li+}$ denotes the surface overpotential of lithium intercalation reaction. When $\eta_{Li|Li+} < 0$ V, lithium ions start to deposit on the graphite surface. One can extract the value of *U* and $|\eta_{LixC6|Li+}|$ from the numerical model and then determine whether lithium plating will happen.

$$\eta_{Li|Li_{+}} = \Phi_s - \Phi_e - iR_f = U - \left| \eta_{LixC6|Li_{+}} \right|$$
(17)

It should be noted that the reaction is nonuniform across the thickness of the electrode; as shown in Fig. S4, the minimum value of *U* and the maximum value of $|\eta|_{LixC6|Li+}|$ always occur at the negative-electrode separator interface (NSI). In other words, lithium plating is most likely to happen near NSI [66]. The maximum reaction rate occurs at NSI since the resistance of the current flow is much smaller through the solid phase than the electrolyte phase. In the flowing analysis, we will use the numerical results on NSI to study lithium plating during fast charging under four different conditions:

- A. Fast charging a partially charged baseline cell (36% initial SOC)
- B. Fast charging a partially charged cell (initial 47% SOC) with ATM method
- C. Fast charging a baseline cell with low initial SOC (5% SOC)

D. Fast charging a cell with low initial SOC (5% SOC) with ATM method

Fig. 7a and 7b demonstrate the potential evolution at the reaction surface of case-A and case-B, respectively; the cases are extracted from the charging step of the UAM cycles. A large surface overpotential of lithium intercalation reaction is the main reason for lithium plating in case-A. Noting that the activation energy of the reaction is relatively high (65 kJ/mol [67], making i₀ increase ~16× from 25 °C to 60 °C), an effective way to reduce the overpotential is to elevate the cell temperature. With the preheating step, the surface overpotential in case-B is notably reduced. According to Eq. (17), a small U value at the electrochemical interface is also prone to leading to negative η_{LilLi+} . Compared with the equilibrium state, the value of U during charging is always smaller as caused by limited ion transport through the electrolyte and solid particles. The lithium stoichiometry at the graphite surface could approach unity, even though the SOC for the cell is still low. The horizontal gap between the dash-dot line and the solid line is indicative of the local difference between the average and interfacial SOCs (or equivalently Li stoichiometry) due to limited mass transport. Comparing case-A and case-B, it is found that the mass transport loss is decreased with the preheating step as well.

Fig. 7c and 7d compare simulation results for case-C with case-D; both cells are charged from a lower initial SOC, a situation similar to an XFC scenario. The mass transport loss signified by the gap between the dash-dot line and the solid line in Fig. 7c is more significant than that in Fig. 7a. However, the shaded area in Fig. 7c, indicative of the onset of Li plating, is smaller than that in Fig. 7a, meaning that Li-plating is less severe. This is because the propensity of Li plating is less when the SOC is very low or close to zero. Indeed, in case-A, the cell has higher initial SOC, and the value of U is about 120 mV. Whereas in case-C, the initial value of U is about 200 mV, leaving more buffer space to avoid lithium plating. Once the cell SOC increases with time during charging in case-C, the cell temperature also increases, as shown in Fig. 7e, thus suppressing the tendency of Li plating. For case-D with higher initial SOC, the ATM cell is at the elevated temperature around 60 °C prior to charging, as depicted in Fig. 7f, and so it exhibits enhanced kinetic and transport properties during fast charging, which avoids Li plating. In short, the ATM cell is safe from lithium plating for both low and high initial SOC, another crucial attribute for UAM applications.

5.3. SOC window of LiB under UAM cycling

Under low charge and discharge rates, the LiBs usually cycles between 0% to 100% SOC to store and release maximum energy. However, owing to the large polarization originates from the sustained high-power input, the maximum capacity of LiBs is usually less than the rated capacity after fast charging. Similarly, since the battery must support the high-power demand for landing and a diverted flight at the end of discharge, we need to store 15-20% capacity to avoid insufficient power output [7]. Voltage loss at the end of charge (EOC) and the end of discharge (EOD) reduces the usable energy of the battery. Low internal resistance is favorable to the widening of the SOC window.

By using the ATM method, the internal resistance of the cell is reduced significantly during charging. According to Fig. 8a, the cell's overpotential is much lower than the baseline case; consequently, the ATM cell could store more energy and sustain a higher voltage during landing. The ATM cell has an end of charge (EOC) SOC of 88%, as compared to 80% for the baseline cell. During discharge, the temperature difference between ATM and baseline cells is minimal. Thus, two cells have similar voltage profiles during discharge. Fig. 8b demonstrates the evolution of current under cycling; the discharge currents are identical for ATM and baseline cells, with 3 C for takeoff, 3.5 C for landing, and around 1 C for cruising. The EOD SOC of ATM and baseline cells are 47% and 36%, respectively.

The SOC window under UAM cycling is an important indicator of battery performance. The maximum SOC relates to the usable energy, and the minimum SOC determines whether the cycling could sustain. With the ECT model, we can predict the SOC window of the battery under the given cycling protocol. Fig. 9a depicts the relationship between initial SOC and EOC SOC. The curve consists of two segments, corresponding to different termination conditions of fast charging. If the initial SOC is lower than 40%, then the charging time will be longer than 5 min, and the charging ends when the charging current is reduced below 3 C. In the other case, the cutoff current drops below 3 C within 5 min; and we would stop charging at 5 min. In both cases, the ATM cell has a higher EOC SOC than the baseline cell. Fig. 9b shows the EOD capacity as a function of initial SOC. The cell voltage drops below 2.7 V when EOD SOC is around 15%, which is consistent with published work [7], and we can set EOD SOC below 15% as the unusable energy. Now that we have the relationship of EOC SOC and EOD SOC vs. initial SOC, we can combine the curves in Fig. 9a and 9b to get the balanced SOC window for batteries. As shown in Fig. 9c and 9d, the SOC window is predicted to be 47~88% for the ATM cell, and 38~81% for the baseline cell, which is very close to the experimental values reported earlier.

5.4. Degradation of LiB under UAM cycling

As the cell degrades, the usable energy of the cell declines, which changes the voltage profile (shown in Fig. S5) and the SOC window during cycling. There are two degradation parameters of critical importance in UAM application: capacity retention and resistance rise. The capacity loss comes from the formation and growth of solid-electrolyte-interphase (SEI) and/or lithium plating, both of which consume cyclable lithium in the cell. The remaining capacity is crucial in determining the cruising range and the payload of an eVTOL [7]. The capacity retention of 80% marks the end of life for a LiB. Additionally, there is a rise in the internal resistance as LiB ages. The UAM application involves high-rate charge and discharge, which makes the internal resistance another critical factor to affect the performance of the aged cells. In this section, we will use the model to simulate UAM cells under various degradation conditions. By doing so, we are able to describe the change of useable capacity during UAM cycles and find out whether a LiB with 80% capacity retention is still suitable for the electric VTOLs.

Fig. 10a shows the evolution of the end-of-charge (EOC) SOC. All the data in the figure are normalized with the rated capacity of the fresh cell. The ATM cell not only has a longer cycle life but also possesses higher SOC at EOC. Fig. 10b demonstrates the change of the end-of-discharge (EOD) SOC with cycle number. Since the discharge energy is fixed for every cycle, there is only a trivial change in discharge capacity; the curves in Fig. 10a and 10b have similar shapes. EOC and EOD SOC values are shown in Fig. 10a, 10b indicate the upper and lower bounds of SOC during UAM cycling and how the SOC range varies with cycle number.

The experimental results of EOC SOC vs. capacity retention are shown in Fig. 10c as symbols for both ATM and baseline cells. To study the aging behavior, we simulate the LiBs at different degradation states by adjusting the parameters in the ECT model. Specifically, we adjust the Bruggeman factor (Eq. (9)) and the exchange current density (Eq. (6)) to account for the increase of ohmic resistance and charge transfer resistance, respectively, upon aging. Furthermore, we adjust the amount of reversible lithium to account



Fig. 10. Variations of end-of-charge (EOC) SOC and end-of-discharge (EOD) SOC under UAM cycling. a) Evolution of EOC SOC. b) Evolution of EOD SOC. c) Relationship of EOC SOC with capacity retention. Symbols: experimental data. Solid lines: model results considering capacity loss only. Dashed lines: model results considering capacity loss and resistance doubling at the end of life (i.e., 20% capacity loss). d) Relationship of EOD SOC with capacity retention. Due to high power discharge during landing, there is about 15% unusable capacity during the UAM cycles.

for the capacity loss. Detailed model parameters for the aged cell are summarized in Tables S3 and S4.

After modeling the degraded cells, we can use the method presented in the previous section to calculate the SOC window at each degradation state. Predicted results are added as solid lines, considering the degradation mechanism only involving lithium loss without a change in the internal resistance. The numerical model predicts a decrease of EOC SOC from 88% to 70% for the ATM cell, and from 81% to 64% for the baseline cell when reaching 80% capacity retention. If accounting for the doubling internal resistance of the cell at EOL due to degradation, the predictions displayed in the dashed lines show that both ATM and baseline cells lose about 6% capacity from the growth of internal resistance.

The EOD capacity, or reserved capacity, is shown in Fig. 10d. Due to high power discharge steps at the end of the cycle, not all of the reserved capacity can be used up. The EOD capacity of the baseline cell is about 10% when the minimum voltage drops below 2.7 V during cycling, which implies it is necessary to keep the EOD capacity above 15% for UAM cycles. We can calculate the usable capacity after discharge by subtracting the unusable capacity from EOD capacity, which could be converted to an extra range or cruising time with simple calculations. According to Fig. 8b, the discharge power for the fixed-wing cruise is 0.9 W/Wh, which means the discharge rate is around 0.9 C. Assume the battery discharge at 1 C during the cruise; every 10% of the rated capacity could transform into 6 min of extra cruise time or 15 miles of extra travel distance.

It can be seen from Fig. 10c and 10d that the experimental data of the ATM and baseline cells both follow the predictions based on

the degradation mode of both capacity loss and internal resistance doubling. Thus, one may extrapolate from the predicted result (i.e., the dashed line) that for an ATM cell at its EOL, there is still about 11% usable capacity after discharge, equivalent to 7 min of extra cruise time or 17 miles of extra travel distance.

6. Conclusions

We have demonstrated a LiB of 209 Wh/kg for short-range UAM applications. This level of energy density is consistent with commercially available energy-dense BEV batteries and lends feasibility to the first generation of eVTOLs for short-range applications. By applying the ATM method, we have shown 5-min charging of this battery simultaneously with long cycle life. The ultrafast charging capability lends a possibility for a high usage rate of eVTOLs and hence their commercial viability. Both upper and lower bounds of SOC during UAM load cycling are suitable for eVTOL maneuver and safety. Our experimental results show that the cell has only 2.3% capacity loss after 1,000 UAM cycles. This cycle life is sufficient for eVTOL applications.

We showed, both experimentally and theoretically, that the ATM method, i.e., preheating a LiB cell to ~60 °C prior to fast charging, can effectively alleviate Li plating, which is a major scientific challenge facing UAM applications. More importantly, we discovered that the ATM method works equally well for both low and high initial SOCs of LIB cells, which is a particularly important attribute for UAM batteries because of their high initial SOC prior to charging required by safety regulations.

Future work consists of extending the cell's specific energy

which could relax the payload restriction to only 100~300 kg with a 140-kWh battery pack. The specific energy can be increased by increasing the areal loading of active materials (e.g., cathode areal capacity to 3~4 mAh/cm²) and by using active materials with higher specific capacities (e.g., Ni-rich cathodes, silicon or silicon-carbon anodes). We shall address these issues in future publications.

CRediT authorship contribution statement

Teng Liu: Conceptualization, Investigation, Writing - original draft. Xiao-Guang Yang: Methodology, Writing - review & editing. Shanhai Ge: Resources. Yongjun Leng: Resources. Chao-Yang Wang: Supervision, Conceptualization, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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