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Nickel-Based Cathode Materials for Lithium-Ion Batteries

1. Introduction

Lithium-ion batteries were first used in practical applications in the 1990s, and have now become an indispensable presence in everyday life as a power source for cellphones, notebook-type personal computers and other mobile devices. The battery capacity of the cylindrical 18650 type, which is mainly used as a power source for power tools, has increased from 1.0 Ah when first marketed to 3.0 Ah or more at present. This high capacity was achieved as a result of improvements in the cathode and anode materials, electrolyte, separator and other component parts. Realizing higher capacity cathode materials will be a key challenge for utilizing this type of lithium-ion battery as a power source for electric vehicles (EV) and energy storage systems (ESS), in which even higher capacities will be necessary.

Lithium cobalt oxide (LCO) and lithium nickel cobalt manganese oxide (NCM tertiary system) have mainly been used as cathode materials. However, in recent years, NCM with a higher compound ratio of Ni and lithium nickel cobalt aluminum (NCA) have attracted interest from the viewpoint of higher capacity.

As a major product that features high capacity, high safety and high cycling stability, JFE MINERAL supplies the NCA-based material 503LP to a wide range of fields, including electric power tools, vacuum cleaners and other power tools, digital cameras, unmanned aerial vehicles (drones), etc., contributing to high performance in those products. In order to respond to the need for even higher capacity of recent years, we are also devoting great effort to the development of a NCA cathode material with a higher compound ratios of Ni.

2. Battery Performance of 503LP

Figure 1 shows the charge and discharge properties of 503LP measured under a 25°C atmosphere using a simple battery cell assembled using metallic lithium (Li) in the counter electrode and 1M-$LiPF_6$/EC+DMC in the electrolyte. The charge property was measured under the conditions of 0.1 C and 4.3 V-CCCV, and the discharge property was measured under 0.1 C and 3.0 V cut-off. The discharge capacity was 188 mAh/g, and charge-discharge efficiency was 89.2%. A high capacity in comparison with LCO and NCM111 ($LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$), etc. is a distinctive feature of this product.

Table 1 shows the specification of a protype laminated battery for evaluation of cycle performance, which represents cycle life performance, and safety. 503LP is used in the cathode, and general materials are used in the other components. Volumetric energy density is 370 Wh/L.

Figure 2 shows the results of the cycle performance evaluation. Cycle performance was evaluated under the measurement conditions of voltages in the range of

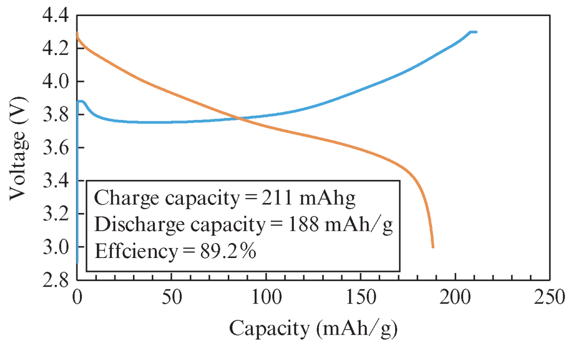

Fig. 1 Charge and discharge properties of 503LP

Table 1 Specification of prototype cell

Cell	Packaging	Aluminum laminated type
	Size	124 × 72 × ᵗ2.3 mm
	Volume	20.5 cm^3
Cathode	Product name	503LP
	Compound ratio	Cathode: C: PVDF = 95: 2: 3
	Current collector	Aluminum (ᵗ15 µm)
Anode	Product	MCMB graphite
	Compound ratio	Anode: C: PVDF = 92.5: 0.5: 7
	Current collector	Copper (ᵗ10 µm)
Separater		Celgard 2325 (ᵗ25 µm)
Electrolyte		1M $LiPF_6$/PC+EC+DEC
Battrey capacity		2.1 Ah

† Originally published in *JFE GIHO* No. 47 (Feb. 2021), p. 66–68

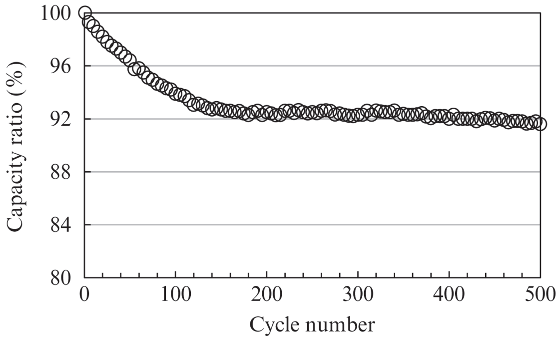

Fig. 2 Cycle performance of 503LP

Table 2 Safety test results

Cathode material	Overcharge test		Nail penetration test	
	Cell temperature (˚C)	Evaluation	Cell temperature (˚C)	Evaluation
503LP	42	Good	155	Good
NCM	56	Good	258	Good

4.2 V to 2.75 V, current of 1 C and room temperature. After 200 cycles, the curve becomes flat, indicating high cycle performance. The discharge capacity retention ratio at 500 cycles was 92%.

Table 2 shows the results of the safety evaluation. In the overcharge test, the battery was charged in advance to 4.2 V, and then charged from that condition to 12 V at a 1 C current value and held for 5 minutes, and the occurrence/nonoccurrence of ignition and the surface temperature of the cell were evaluated. In the Nail Penetration test, a stainless steel nail with a diameter of 5 mm was driven through the center of the cell while in the 4.2 V fully charged state, and ignition and the surface temperature of the cell were evaluated. In the safety test, a comparison battery with the specification in Table 1 was prepared using NCM111 (high safety cathode material) in place of 503LP. The battery capacity of the NCM111 battery was 1.75 Ah.

The results of this safety test demonstrated that the 503LP battery could secure safety equal or superior to that of the NCM battery.

3. High-Ni Cathode Material 721NT

When the current main stream cathode material NCM was first marketed, its Ni ratio was 0.33 mol (Ni/Metal mol ratio, Metal = Ni, Co, Al, Mn, etc.). However, this has now been increased to Ni ratio=0.50 mol or Ni ratio=0.60 mol in order to achieve higher capacity and reduce the use of expensive Co, and energetic development of new high capacity products using even higher Ni ratios is underway. Although the Ni ratio of 503LP is 0.78 mol, JFE MINERAL is developing a high capacity NCA with a higher Ni compounding ratio to respond to the recent need for higher capacity and differentiate the new product from the conventional NCM.

503EF, which was developed as a high capacity version of 503LP, has a Ni ratio=0.82 mol and a discharge capacity of 196 mAh/g. Customers' certification work has been completed, and the changeover from 503LP to 503EF is progressing, mainly in power tool applications. Although the capacities requested by customers differ depending on the application, in EV applications, a discharge capacity of 210 mAh/g or higher is required in many cases.

If the Ni compounding ratio is increased, only the discharge capacity increases, but in particular, charge/discharge efficiency and cycle performance tend to deteriorate remarkably. For this reason, it is necessary to increase the Ni compound ratio and optimize the composition ratios of other elements and synthesis conditions. In the newly-developed product 721NT, the composition ratio and synthesis conditions were optimized at a Ni ratio=0.87 mol. As shown in **Fig. 3**, this product achieves a discharge capacity of 210 mAh/g and charge-discharge efficiency of 89.3%. **Figure 4** shows a comparison of the cycle performance of 721NT and 503LP when coin cells were prepared using metallic Li as the counter electrode and the cells were evaluated at measurement voltages in the range of

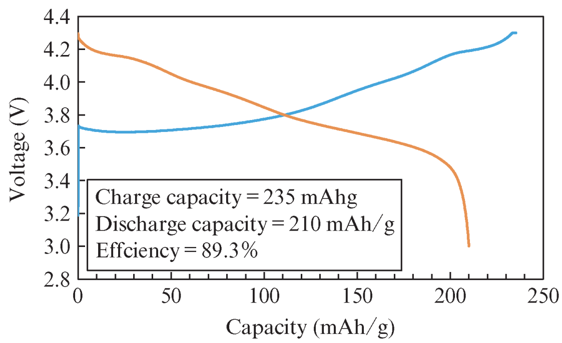

Fig. 3 Charge and discharge properties of 721NT

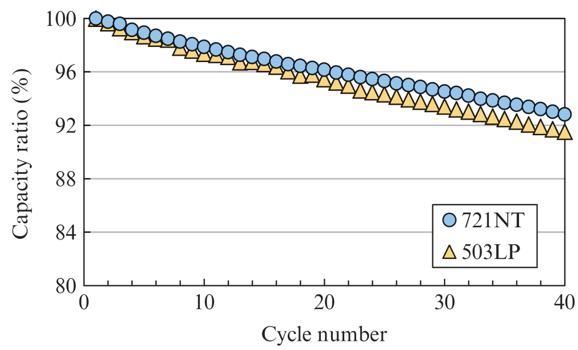

Fig. 4 Cycle performance of 721NT

Table 3 Characteristics of high nickel-based cathode materials

Item			503LP	721NT
Ni/Metal		mol	0.78	0.87
Charge capacity		mAh/g	211	235
Discharge capacity		mAh/g	188	210
Efficiency		%	89.2	89.3
Particle size distribution	D10	µm	8.8	9.3
	D50	µm	13.5	14.2
	D90	µm	19.4	20.0
Compression density		g/cm^3	3.55	3.55
BET Specific surface area		m^2/g	0.55	0.47
Alkali impurities	LiOH	%	0.10	0.13
	Li_2CO_3	%	0.03	0.11

4.25 V to 2.75 V and a 1 C current value under a 25°C environment. The discharge capacity of the new 721NT increased by 10% or more in comparison with the 503LP, and its charge/discharge efficiency and cycle performance were equal or superior to those of the 503LP. The evaluation results for other characteristics were also almost the same as the values for 503LP, as shown in **Table 3**.

4. Conclusion

JFE MINERAL has developed a new high-Ni cathode material product, 721NT, which features high capacity and high cycle performance, and is conducting sample work with the aim of early certification for use in high-grade power tools. We are also developing this product for EV applications, where there is a strong need for batteries that can increase the vehicle travel range per charge.

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