

Application No: GEGADYNE ENERGY LABS PRIVATE LIMITED	Total no. of sheets: 05
Applicant:	Sheet No.: 1

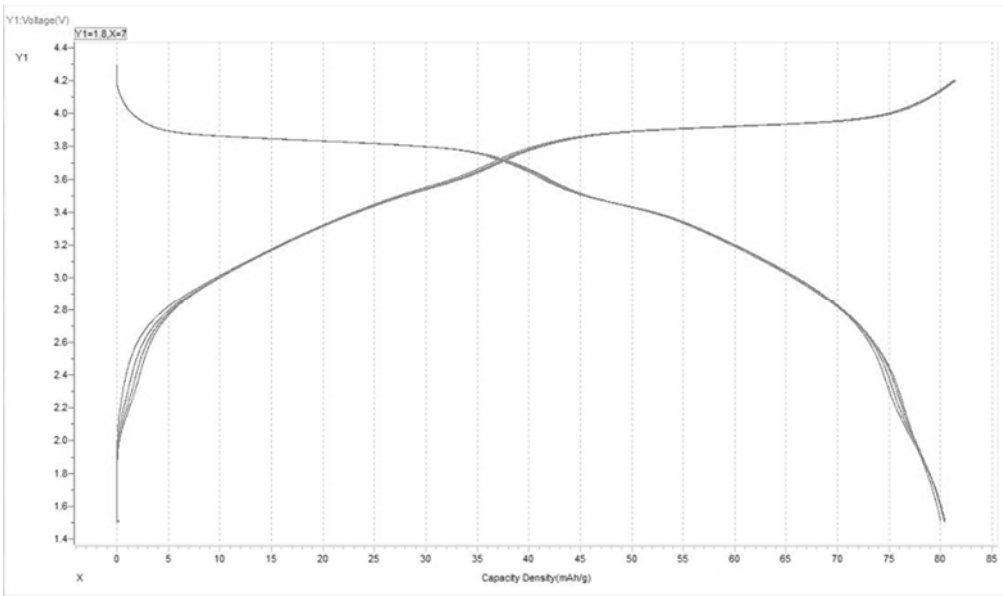


Fig 1 (a)

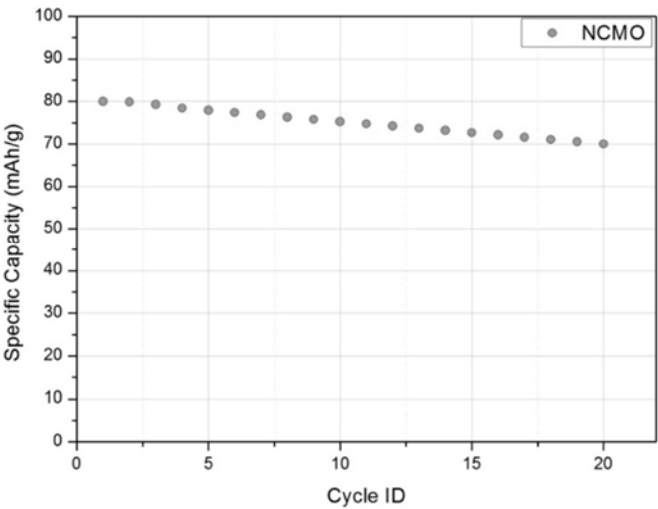


Fig 1(b)

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Application No: GEGADYNE ENERGY LABS PRIVATE LIMITED	Total no. of sheets: 05
Applicant:	Sheet No.: 2

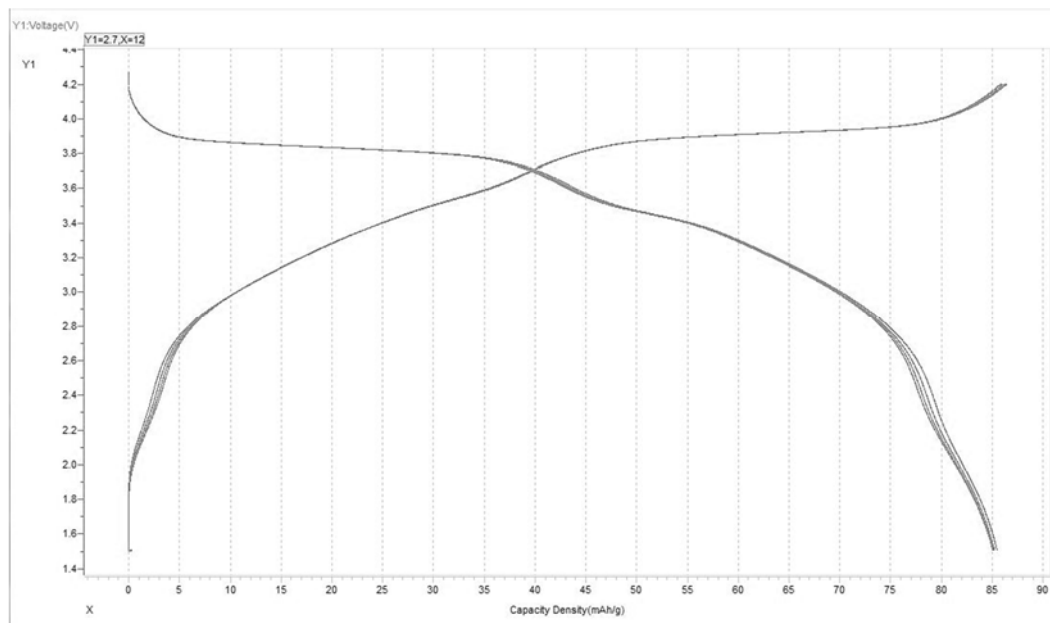


Fig 2(a)

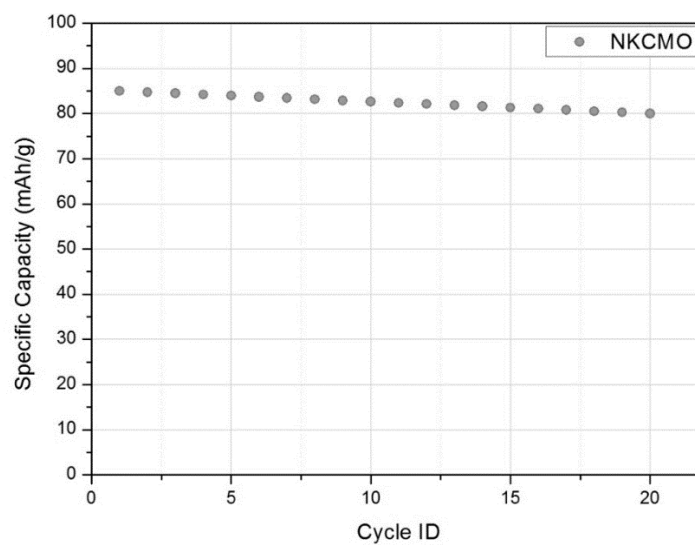


Fig 2(b)

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Applicant:	Sheet No.: 3

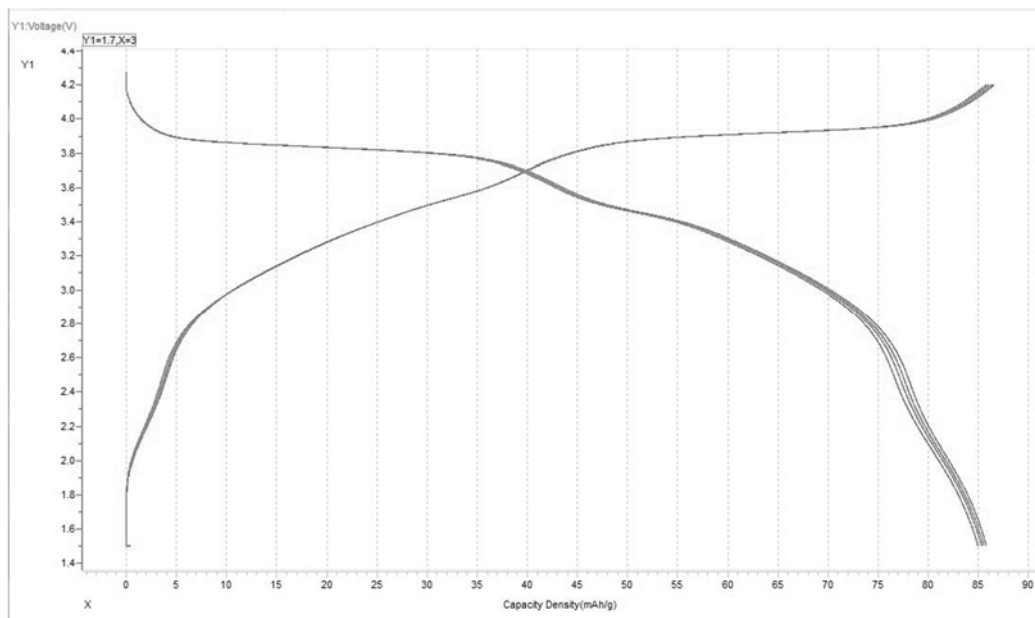


Fig 3(a)

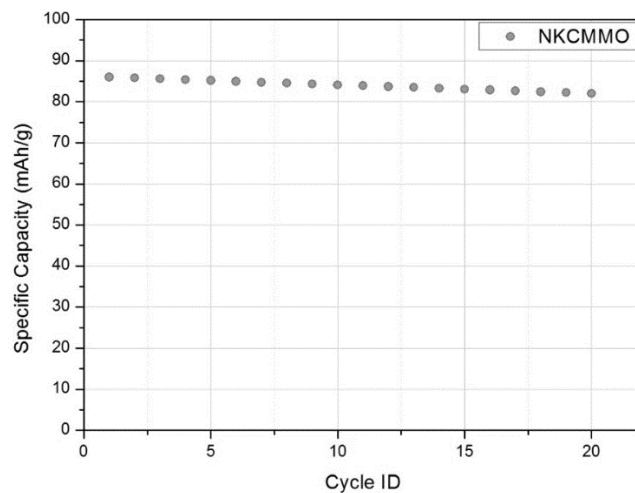


Fig 3(b)

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Application No: GEGADYNE ENERGY LABS PRIVATE LIMITED	Total no. of sheets: 05
Applicant:	Sheet No.: 4

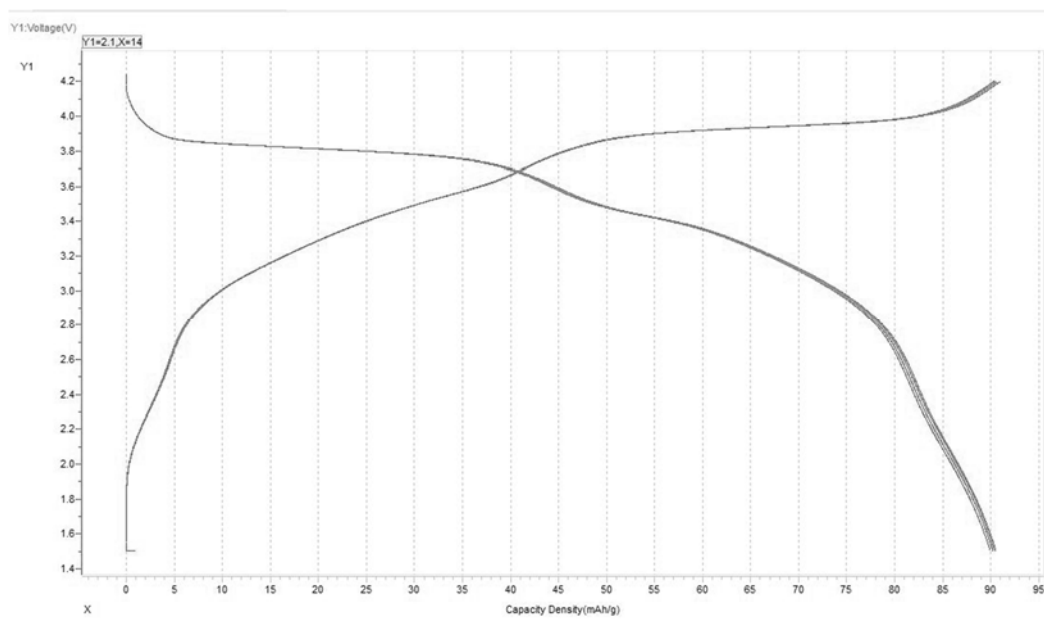


Fig 4(a)

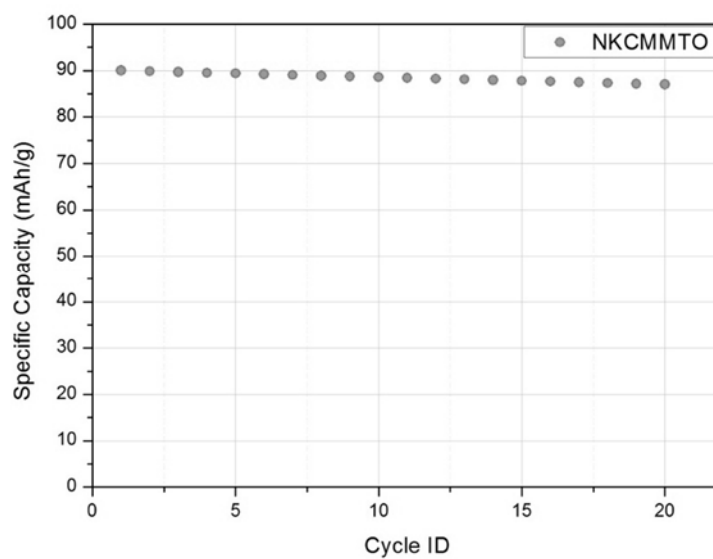


Fig 4(b)

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Applicant:	Sheet No.: 5

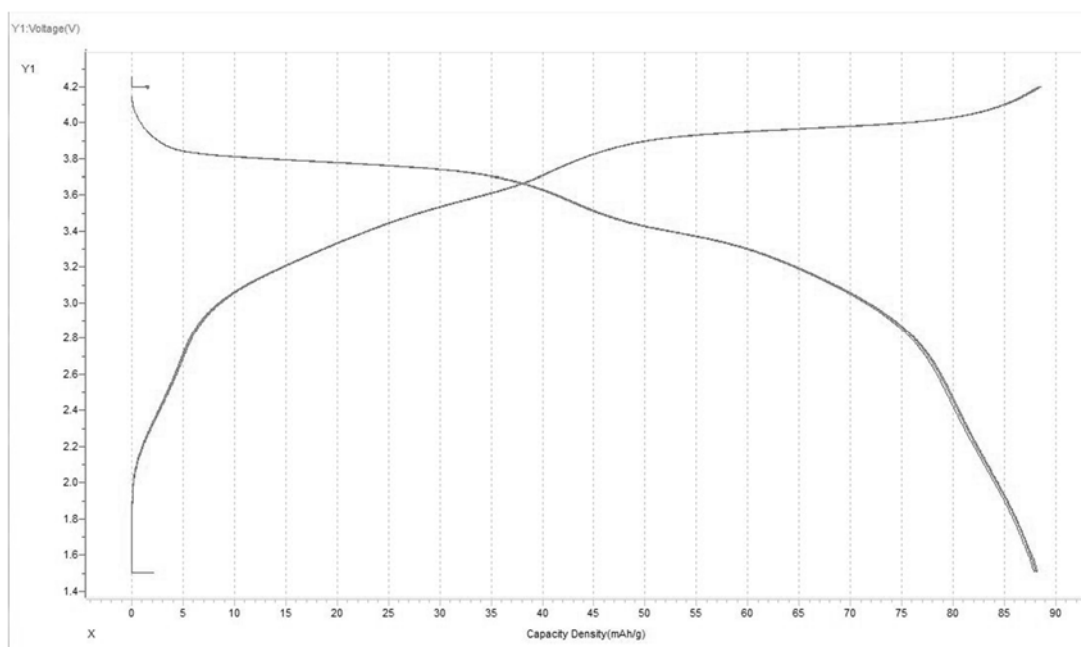


Fig 5(a)

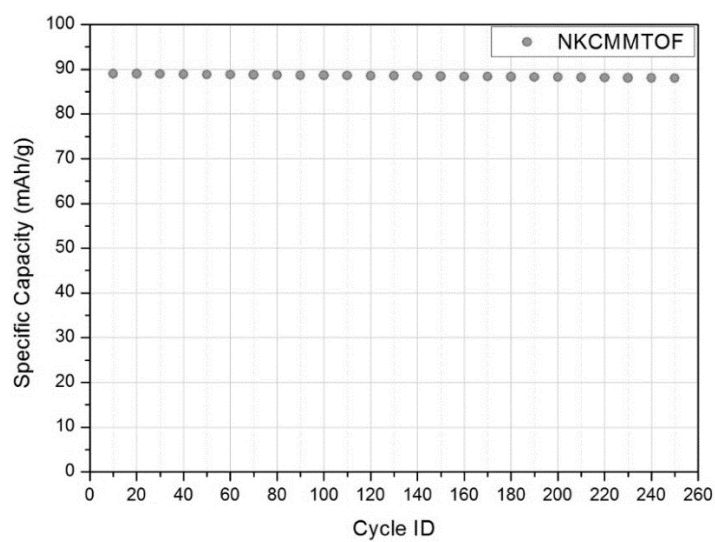


Fig 5(b)

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FORM 2 THE PATENTS ACT, 1970 (39 of 1970) AND The Patents Rules, 2003 COMPLETE SPECIFICATION (See section 10 and rule 13)
1. TITLE OF THE INVENTION: “ MIXED CATION DOPED CUPRATE CATHODES AND PROCESS THEREOF”
2. APPLICANT: (a) NAME: GEGADYNE ENERGY LABS PRIVATE LIMITED (b) NATIONALITY: Indian Company registered under the Companies Act, 2013 (c) ADDRESS: RH32, Pushpanjali, Opp Puranik City, Owala Naka, GB Road, Thane (West) - 400615, Maharashtra, India.
3. PREAMBLE TO THE DESCRIPTION: The following specification particularly describes the invention and the manner in which it is to be performed.

FIELD OF INVENTION:

The present invention relates to mixed cation doped cuprate cathodes for energy devices notably non-aqueous re-chargeable alkali-ion electrochemical cells and batteries and to the process of preparation thereof. More particularly, the present invention relates to doped cuprate cathode active material of Formula (I) that shows little or no fading on cycling, which is able to be recharged multiple times without significant loss in charge capacity.

BACKGROUND & PRIOR ART:

Alkali-ion batteries are analogous in many ways to the lithium-ion batteries that are in common use today; they are both reusable secondary batteries that comprise an anode (negative electrode), a cathode (positive electrode) and an electrolyte material, both are capable of storing energy, and they both charge and discharge via a similar reaction mechanism. When an alkali-ion (or lithium-ion) battery is charging, M^+ (or Li^+) ions de-intercalate from the cathode and insert into the anode. Meanwhile charge balancing electrons pass from the cathode through the external circuit containing the charger and into the anode of the battery. During discharge the same process occurs but in the opposite direction.

Lithium-ion battery technology has enjoyed a lot of attention in recent years and provides the preferred portable battery for most electronic devices in use today; however lithium is not a cheap metal to source and is considered too expensive for use in large scale applications. By contrast, alternative alkali-ion based battery technology is still in its relative infancy but is seen as advantageous; metals like sodium and potassium are much more abundant than lithium and some researchers predict this will provide a cheaper and more durable way to store energy into the future, particularly for large scale applications such as storing energy on the electrical grid.

Metal oxides with the general formula A_xMO_2 (where A represents one or more alkali metal ions and M represents one or more metal ions at least one of which

has several oxidation states, for example a transition metal) are known to crystallise in a number of different layered structures. This is described in detail by C. Delmas et al in "Structural Classification and Properties of the Layered Oxides", Physica 99B (1980) 81-85. In summary, the structures are all made up of MO₆ edge sharing octahedra which form (MO₂)_n sheets. These sheets are stacked one on top the other and are separated by the alkali metal atoms and the exact position of the alkali metal will dictate whether the overall structure of the metal oxide is to be described as octahedral (O), tetrahedral (T) or prismatic (P). In a lattice made up of hexagonal sheets, there are three possible positions for the oxygen atoms, conventionally named A, B and C. It is the order in which these sheets are packed together that leads to the O, T and P environments. The number 2 or 3 is also used to describe the number of alkali metal layers in the repeat unit perpendicular to the layering. For example, when the layers are packed in the order ABCABC, an O₃ structure is obtained. This translates to 3 alkali metal layers in the repeat unit and each alkali metal being in an octahedral environment. Such materials are characterised by the alkali metal ions being in octahedral orientation and typical compounds of this structure are A_xMO₂ (x≤1). The order ABAB with the alkali metal ions in tetrahedral orientation will yield a T₁ structure which is typified by A₂MO₂ compounds. Packing the sheets in ABBA order gives a P₂ structure in which one half of the prism shares edges with MO₆ octahedra and the other half shares the faces. Packing in ABBCCA order results in a P₃ structure type in which all prisms share one face with one MO₆ octahedron and three edges with three MO₆ octahedra of the next sheet and the compounds are found to adopt the P₃ structure. It will be noted that the amount of alkali metal present in the A_xMO₂ material has a direct bearing on the overall structure of the metal oxide.

Attempts are made in the art to dope alkaline batteries or lithium ion batteries with active materials selected from alkali metals, transition metals, lanthanides to increase depth of discharge and to improve the cyclability.

US8475959B2 relates to a positive electrode active material for a lithium ion battery comprising a layered lithium doped lithium metal oxide composition approximately represented by a formula $x\text{Li}_2\text{MnO}_3 \cdot (1-x)\text{LiNi}_{u+\Delta}\text{Mn}_{u-\Delta-d}\text{Li}_d\text{Co}_w\text{A}_y\text{O}_2$, x ranges from about 0.03 to about 0.55, d ranges from about 0.004 to about 0.05, $2u+w+y$ is approximately equal to 1, Δ ranges from about -0.0019 to about 0.0019, w ranges from about 0.2 to about 0.475, u ranges from about 0.2 to about 0.4, y ranges from 0 to about 0.1 with the proviso that both $(u+\Delta)$ and w are not both 0 and A is Mg, Sr, Ba, Cd, Zn, Al, Ga, B, Zr, Ti, Ca, Ce, Y, Nb, Cr, Fe, V, or combinations thereof.

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Article titled “A Manganese-Doped Barium Carbonate Cathode for Alkaline Batteries’ by Benjamin Hertzberg et.al published in Journal of The Electrochemical Society, **161** (6) A835-A840 (2014) discloses substitution of manganese in the Witherite crystal structure of BaCO_3 to provide $\text{BaMn}_{0.51}\text{Co}_{0.49}\text{O}_3$ which creates a high degree of electrochemical activity, superior rate capability and cyclability. The article further provides synthesis of Manganese-doped Witherite via a one-step hydrothermal process.

The article by Su Cheol Han et.al titled ‘Ca-doped Na_xCoO_2 for improved cyclability in sodium ion batteries’ published in Journal of Power Sources **277** (2015) 9e16 provides multi-valent cation substitution for Na^+ as cathode material to improve the cyclability of sodium ion batteries (SIBs) since calcium has no movement in the electrolyte and does not come out of the lattice during charging. The article discloses $\text{Na}_x\text{Ca}_y\text{CoO}_2$ ($0.45 \leq x \leq 0.64$, $0.02 \leq y \leq 0.10$) cathode material synthesized via a solid-state method.

A Ni-doped magnesium manganese oxide as the cathode via a conventional hydrothermal method, which help to improve conductivity and solve defects derived from pure manganese oxide with nickel doping is reported in the article titled “Ni-doped magnesium manganese oxide as a cathode and its application in

aqueous magnesium-ion batteries with high rate performance” by Hongyu Zhang et.al in *J. Name.*, 2013, **00**, 1-3.

US8835041B2 discloses an electrode suitable for use as a cathode in a sodium electrochemical cell or battery, the electrode comprising a layered material of formula $\text{Na}_c\text{Li}_d\text{Ni}_e\text{Mn}_f\text{M}_z\text{O}_b$, wherein M comprises one or more metal cation, $0.24 \leq c/b \leq 0.5$, $0 \leq d/b < 0.23$, $0 \leq e/b \leq 0.45$, $0 \leq f/b \leq 0.45$, $0 \leq z/b \leq 0.45$, the combined average oxidation state of the metallic components Na, Li, Ni, Mn, and M is in the range of about 3.9 to 5.2, and b is equal to $(c+d+V_e+X_f+Y_z)/2$, wherein V is the average oxidation state of the Ni, X is the average oxidation state of the Mn, and Y is the average oxidation state of the M; and wherein a combined positive charge of the metallic components is balanced by a combined negative charge of the oxygen anions, the Na is predominately present in a sodium layer, and the Mn, Ni, and M are predominately present in a transition metal layer.

The recent research by Li Yang et.al in Journal of Electronic Materials published in 2018, The Minerals, Metals & Materials Society titled “Effect of Cu Doping on the Structural and Electrochemical Performance of $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ Cathode Materials has demonstrated that the Cu-doped material exhibits better galvanostatic charge/discharge cycling performance arising from an enlarged Li layer spacing and a reduced degree of cation mixing.

The research article titled “ Cu^{2+} Dual-Doped Layer-Tunnel Hybrid $\text{Na}_{0.6}\text{Mn}_{1-x}\text{Cu}_x\text{O}_2$ as a Cathode of Sodium-Ion Battery with Enhanced Structure Stability, Electrochemical Property, and Air Stability” by Ting-Ru Chen et.al published in ACS Appl. Mater. Interfaces 2018, 10, 12, 10147–10156, the $\text{Na}_{0.6}\text{Mn}_{0.9}\text{Cu}_{0.1}\text{O}_2$ cathode with capacity of 96 mA h g^{-1} retained after 250 cycles at 4 C and 85 mA h g^{-1} at 8 C.

Improvements are made in the art to prevent cation mixing, oxygen evolution, and structural collapse in layered cathode materials by doping with anionic elements

such as F, Cl, and S into the oxygen sites of layered cathode materials to effectively enhance the electrochemical performance.

In the article titled “Fluorine-Doped $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ Cathode for High-
5 Performance Lithium-Ion Batteries” by Hyeona Kim et.al published in Energies
2020, 13, 4808; the oxygen atoms in the NCM are replaced with F- ions to
produce a F-doped NCM structure in lithium-ion batteries, $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$ ($x +$
 $y + z = 1$) (NCM) cathode materials. The article provides anionic doping than
cationic doping.

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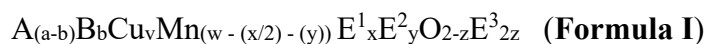
The current workers observed that there is a scope to provide an improved doped
cuprate cathode active material for energy storage devices or alkali-ion
electrochemical cell which is capable of delivering specific capacity performance
with little or no fading on cycling, is able to recharge multiple times without
15 significant loss in charge capacity showing excellent rate performance. This
remains the objective of the present invention.

SUMMARY OF INVENTION:

The primary objective of the present invention is to provide mixed cation doped
20 positive electrodes for energy devices with little or no fading on cycling without
significant loss in charge capacity showing excellent rate performance over the
art. The said effect is achieved in the present invention due to synergistic doping
of the cations and anions.

25 Accordingly, the present invention relates to mixed cation doped positive
electrodes for energy devices that show high specific capacity performance with
little or no fading on cycling.

In an aspect, the present invention provides mixed cation doped cuprate cathode
30 active material of formula (I) for energy devices with little or no fading on
cycling, comprising;



wherein

‘A’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

- 5 ‘B’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

Cu is copper in oxidation state 2+,

Mn is manganese in oxidation state 4+,

E^1 comprises one or more elements in oxidation state 2+,

- 10 E^2 comprises one or more elements in oxidation state 4+, and

E^3 comprises one or more elements in oxidation state 1-

wherein

$0.4 \leq a \leq 0.9$, preferably $0.5 \leq a \leq 0.85$, further preferably $0.6 \leq a \leq 0.75$,

$0.001 \leq b \leq 0.4$, preferably $0.005 \leq b \leq 0.1$, further preferably $0.005 \leq b \leq 0.04$,

- 15 $0 \leq v \leq 0.5$, preferably $0 \leq v \leq 0.45$ and ideally $0 \leq v \leq 0.333$,

$0.4 \leq w \leq 0.8$, preferably $0.45 \leq w \leq 0.7$ and ideally $0.5 \leq w \leq 0.7$,

at least one of x and y is in the range of 0 to 0.39,

z is in the range of 0 to 1.99.

- 20 In another aspect, the doped cuprate cathode active material of Formula (I) comprises;

(i) $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.571}O_2$,

(ii) $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.567}Mg_{0.006}O_2$,

(iii) $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.564}Mg_{0.006}Ti_{0.006}O_2$,

- 25 (iv) $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.564}Mg_{0.006}Ti_{0.006}O_{1.99}F_{0.02}$,

In an aspect, the mixed cation doped cuprate based cathode active material with optimized stoichiometry is provided such that the P1 structure of the formed cuprate is doped with larger alkali ions and results in enlarged interlayer spacing, providing larger channels for ion movement. The +2 and +4 doped elements also reinforce the manganese back-bone and prohibits phase transitions which

30

otherwise would lead to degradation over multiple cycles. Along with cation doping, the current invention also utilizes anionic doping with element having -1 oxidation state to further increase strength of the bonds and reduce oxygen loss at high potentials.

5

In yet another aspect, the cathode active material of the present invention may be prepared by any known and/or convenient process. Accordingly, one or more precursor material comprising one or more elements selected from A, B, E¹, E², and E³(as defined above) in a stoichiometric ratio that corresponds with the amounts of the respective one or more metals present in said Active Material may be heated (for example in a furnace) in order to facilitate a solid state reaction process. Such a process may be conveniently performed in the presence of air, but it may also be performed under an inert atmosphere.

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15 In an aspect, the active material of Formula (I) is used as an electrode preferably a positive electrode (cathode), and further preferably in conjunction with a counter electrode and one or more electrolyte materials in alkali ion-cell and in energy storage devices or electrochemical cell.

20 In another aspect, the present invention provides alkali-ion electrochemical cell comprising mixed cation doped cuprate cathode active material with little or no fading on cycling of the formula (I);



25 wherein

‘A’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

‘B’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

30 Cu is copper in oxidation state 2+,

Mn is manganese in oxidation state 4+,

E^1 comprises one or more elements in oxidation state 2+,
 E^2 comprises one or more elements in oxidation state 4+, and
 E^3 comprises one or more elements in oxidation state 1-
wherein

- 5 $0.4 \leq a \leq 0.9$, preferably $0.5 \leq a \leq 0.85$, further preferably $0.6 \leq a \leq 0.75$,
 $0.001 \leq b \leq 0.4$, preferably $0.005 \leq b \leq 0.1$, further preferably $0.005 \leq b \leq 0.04$,
 $0 \leq v \leq 0.5$, preferably $0 \leq v \leq 0.45$ and ideally $0 \leq v \leq 0.333$,
 $0.4 \leq w \leq 0.8$, preferably $0.45 \leq w \leq 0.7$ and ideally $0.5 \leq w \leq 0.7$,
at least one of x and y is in the range of 0 to 0.39,
10 z is in the range of 0 to 1.99.

In an aspect, the present invention provides electrode active material of Formula (I) in conjunction with a counter electrode and one or more electrolyte materials in energy storage devices.

15

In another aspect, the charge-discharge profile of the present electrode active material of Formula (I) exhibits two plateaus at about 4.0 V and 3.5V when used as the cathode in energy storage devices comprising hard carbon as anode and the electrolyte comprising of 0.5M solution of NaPF_6 in a mixture of Ethylene
20 Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1.

In another aspect, the present invention provides an alkali-ion electrochemical cell comprising,

25

- (i) An anode;
- (ii) A cathode containing the doped cuprate active material of Formula (I);
- (iii) A separator between the positive electrode and negative electrode; and
- (iv) An electrolyte.

The anode comprises the hard carbon and the electrolyte comprises 0.5 M solution of NaPF₆ and 0.3M solution of KPF₆ in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1.

- 5 The mixed cation doped cuprate cathode active material of Formula (I) of the present invention find application in alkali ion-cell, in energy storage devices such as batteries, rechargeable batteries, electrochemical devices and electrochromic devices.

10 **BRIEF DESCRIPTION OF DRAWINGS:**

FIGURE 1 (a) shows the cell voltage profile for the first 5 charge/discharge cycles of the cell having hard carbon as anode material and Na_{0.657}Cu_{0.314}Mn_{0.571}O₂ as cathode active material;

- 15 **FIGURE 1 (b)** shows the Constant current cycling (CC/CV) of a full cell with hard carbon as anode material and Na_{0.657}Cu_{0.314}Mn_{0.571}O₂ in the voltage range 1.5- 4.2V at 25°C in 0.5M NaPF₆ in EC:DEC:FEC and PP used as a separator;

FIGURE 2 (a) shows the cell voltage profile for the first 5 charge/discharge cyclings of the cell having Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.571}O₂ as cathode active material;

- 20 **FIGURE 2 (b)** shows the constant current cycling (CC/CV) of a cell with Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.571}O₂ as cathode active material in the voltage range 1.5- 4.2V at 25°C in 0.5M NaPF₆ and 0.3M KPF₆ in EC:DEC:FEC and PP used as a separator;

- 25 **FIGURE 3 (a)** shows the cell voltage profile for the first 5 charge/discharge cycles of the cell having Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.567}Mg_{0.006}O₂ cell;

- FIGURE 3 (b)** shows the constant current cycling (CC/CV) of a cell having Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.567}Mg_{0.006}O₂ as cathode active material in the voltage range 1.5- 4.2V at 25°C in 0.5M NaPF₆ and 0.3M KPF₆ in EC:DEC:FEC and PP used as a separator;
- 30

FIGURE 4 (a) shows the cell voltage profile for the first five charge/discharge cycles of the cell having $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_2$ as cathode active material;

FIGURE 4 (b) shows the constant current cycling (CC/CV) of a cell having $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_2$ as cathode active material in the voltage range 1.5- 4.2V at 25°C in 0.5M NaPF6 and 0.3M KPF6 in EC:DEC:FEC and PP used as a separator;

FIGURE 5 (a) shows the cell voltage profile for the first five charge/discharge cycles of $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_{1.99}\text{F}_{0.02}$ as cathode active material;

FIGURE 5 (b) shows the constant current cycling (CC/CV) of the cell with $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_{1.99}\text{F}_{0.02}$ as cathode active material in the voltage range 1.5- 4.2V at 25°C in 0.5M NaPF6 and 0.3M KPF6 in EC:DEC:FEC and PP used as a separator;

DETAILED DESCRIPTION OF INVENTION:

The features of the present invention is described below in the various preferred and optional embodiments, however, should not be construed as limiting the scope of the invention.

The present invention relates to cation doped cuprate cathode active material which is cost effective in comparison to Li-ion batteries. The cathode active material of the present invention can be easily synthesized, exhibit high rates, high energy, and long cycle life in energy storage devices with little or no fading on cycling. The inventive feature of the present invention is the mixed cation doping of the cuprate cathode wherein the larger doped cations results in enlarged interlayer spacing, providing larger channels for ion movement thereby increasing the retention of capacity across multiple cycles. Along with doped alkali ions of larger size, the non-participating +2 and +4 ions reinforce the manganese backbone preventing the phase transitions during charging and in a preferred

aspect of the invention the monovalent anionic doping prevents the cation mixing and oxygen evolution.

In an embodiment, the present invention discloses mixed cation doped cuprate cathode active material, with little or no fading on cycling of the formula (I) for energy devices comprising,



wherein

‘A’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

‘B’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

Cu is copper in oxidation state 2+,

Mn is manganese in oxidation state 4+,

E¹ comprises one or more elements in oxidation state 2+,

E² comprises one or more elements in oxidation state 4+, and

E³ comprises one or more elements in oxidation state 1-

wherein

0.4 ≤ a ≤ 0.9, preferably 0.5 ≤ a ≤ 0.85, further preferably 0.6 ≤ a ≤ 0.75,

0.001 ≤ b ≤ 0.4, preferably 0.005 ≤ b ≤ 0.1, further preferably 0.005 ≤ b ≤ 0.04,

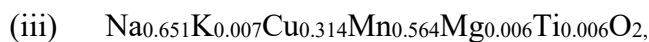
0 ≤ v ≤ 0.5, preferably 0 ≤ v ≤ 0.45 and ideally 0 ≤ v ≤ 0.333,

0.4 ≤ w ≤ 0.8, preferably 0.45 ≤ w ≤ 0.7 and ideally 0.5 ≤ w ≤ 0.7,

at least one of x and y is in the range of 0 to 0.39,

z is in the range of 0 to 1.99.

In another embodiment, the mixed cation doped cathode active material of Formula (I) comprises;



In an embodiment, the present invention discloses a mixed cation doped cuprate based cathode active material with optimized stoichiometry such that the P1 structure of the formed cuprate is doped with larger alkali ions and results in enlarged interlayer spacing, providing larger channels for ion movement. The +2 and +4 doped elements also reinforce the manganese back-bone and prohibits phase transitions which otherwise would lead to degradation over multiple cycles. Along with cation doping, the current invention also utilizes anionic doping with a monovalent fluorine atom to further increase the strength of the bonds and reduce oxygen loss at high potentials.

10

In an embodiment, the present invention discloses electrode active material of Formula (I) in conjunction with a counter electrode and one or more electrolytes in energy storage devices.

15 In another embodiment, the charge-discharge profile of the present electrode active material of Formula (I) exhibits two plateaus at about 4.0 V and 3.5V when used as the cathode in energy storage devices comprising hard carbon as anode and the electrolyte comprising of 0.5M solution of NaPF₆ in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1.

20

In another embodiment, the present invention discloses a method for preparation of said cathode active material of Formula (I) comprising;

- 25 (i) Intimately mixing together the starting materials (i.e. the precursors for the Cathode Active Material) in the correct stoichiometric ratio and pressing into a pellet;
- (ii) Heating the resulting mixture in a furnace under a suitable atmosphere comprising ambient air, or an inert atmosphere of argon or nitrogen, wherein the gases may be flowing if desired, at a single temperature or over a range of temperatures between 450°C and 800°C until reaction product forms; and optionally

30

(iii)Allowing the product to cool before grinding it to a powder.

The Table 1 below lists the starting materials and heating conditions used to prepare a known (comparative) composition (Example 1) and the Target Active
5 Materials of the present invention (Examples 2 to 5).

Table 1:

1	$\text{Na}_{0.657}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$	0.657 NaNO_3 , 0.314 CuO , 0.571 MnCO_3	600°C, Oxygen Dwell: 4 Hrs Ramp Rate: 10°C/min.
2	$\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$	0.651 NaNO_3 , 0.007 KNO_3 , 0.314 CuO , 0.571 MnCO_3	600°C, Oxygen Dwell: 4 Hrs Ramp Rate: 10°C/min
3	$\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.567}\text{Mg}_{0.006}\text{O}_2$	0.651 NaNO_3 , 0.007 KNO_3 , 0.314 CuO , 0.567 MnCO_3 , 0.006 Mg(OH)_2	600°C, Oxygen Dwell: 4 Hrs Ramp Rate: 10°C/min
4	$\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_2$	0.651 NaNO_3 ,	600°C,

		0.007KNO ₃ , 0.314 CuO, 0.564 MnCO ₃ , 0.006Mg(OH) ₂ 0.006 TiO ₂	Oxygen Dwell: 4 Hrs Ramp Rate: 10°C/min
5	Na _{0.651} K _{0.007} Cu _{0.314} Mn _{0.564} Mg _{0.006} Ti _{0.006} O _{1.99} F _{0.02}	0.631 NaNO ₃ , 0.007 KNO ₃ , 0.314 CuO, 0.564 MnCO ₃ , 0.006Mg(OH) ₂ 0.006 TiO ₂ 0.02 NaF	600°C, Oxygen Dwell: 4 Hrs Ramp Rate: 10°C/min

In another embodiment, the present invention discloses the alkali ion-electrochemical cell comprising mixed cation doped cuprate cathode active material, with little or no fading on cycling, of formula (I);

$$5 \quad A_{(a-b)}B_bCu_vMn_{(w-(x/2)-(y))}E^1_xE^2_yO_{2-z}E^3_{2z} \quad \textbf{(Formula I)}$$

wherein

‘A’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

10 ‘B’ comprises one or more alkali metals selected from lithium sodium, potassium and cesium;

Cu is copper in oxidation state 2+,

Mn is manganese in oxidation state 4+,

E¹ comprises one or more elements in oxidation state 2+,

E² comprises one or more elements in oxidation state 4+, and

15 E³ comprises one or more elements in oxidation state 1-

wherein

0.4 ≤ a ≤ 0.9, preferably 0.5 ≤ a ≤ 0.85, further preferably 0.6 ≤ a ≤ 0.75,

0.001 ≤ b ≤ 0.4, preferably 0.005 ≤ b ≤ 0.1, further preferably 0.005 ≤ b ≤ 0.04,

$0 \leq v \leq 0.5$, preferably $0 \leq v \leq 0.45$ and ideally $0 \leq v \leq 0.333$,
 $0.4 \leq w \leq 0.8$, preferably $0.45 \leq w \leq 0.7$ and ideally $0.5 \leq w \leq 0.7$,
 at least one of x and y is in the range of 0 to 0.39,
 z is in the range of 0 to 1.99.

5

In a preferred embodiment, the present invention discloses the cathode active material comprising $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_{1.99}\text{F}_{0.02}$ with capacity fade of about 1.12 % over 250 cycles.

- 10 In yet another embodiment, the present invention provides alkali-ion electrochemical cell comprising,
- (i) An anode;
 - (ii) A cathode containing the doped cuprate active material of Formula (I);
 - (iii) A separator between the positive electrode and negative electrode; and
 - 15 (iv) An electrolyte.

The anode comprises the hard carbon and the electrolyte comprises 0.5 M solution of NaPF_6 and 0.3 M solution of KPF_6 in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1.

20

In an embodiment, the mixed cation doped cuprate cathode active material in said electrochemical cell comprises the formed cuprate doped with larger alkali ions that results in enlarged interlayer spacing, providing larger channels for ion movement; the +2 and +4 doped elements reinforce the manganese back-bone and

25 prohibits phase transitions and the degradation over multiple cycles; the anionic doping with a monovalent atom of 1- oxidation state increases the strength of the bonds and reduces oxygen loss at high potentials.

30

Accordingly, during cell charging process, host ions comprising the larger alkali metal ions are extracted from the cathode active material, and inserted into the Hard Carbon anode, increasing the gallery height of the said carbon anode layers,

thereby helping in unimpeded movement of the smaller host ions, leading to better capacity retention across multiple cycles in the full cell comprising the said cathode, a standard anode, and an electrolyte. The copper ions are oxidized from +2 to +3 state when the cations are deintercalated from the cathode during charging and are responsible for the peaks at ~3.6V and 4.0V. The capacity contribution of Manganese is insignificant and is only seen below 3V as a sloping curve. During the subsequent discharge process, said host ions are extracted from the Hard Carbon and re-inserted into the cathode active material.

10 In an embodiment, the voltage profile of the present electrode material of Formula (I) shows two plateaus at around 3.6V and around 4V, which is smooth and repeatable between voltage cutoffs of about 2.0 to about 4.2V indicating that the active material of Formula (I) retains its single phase structure during the entire charge –discharge cycles.

15

In a preferred embodiment, the present invention discloses the alkali ion-electrochemical cell comprising;

- (i) $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_{1.99}\text{F}_{0.02}$ as cathode active material with little or no fading on cycling;
- 20 (ii) Hard Carbon as anode material;
- (iii) A separator between the positive electrode and negative electrode; and
- (iv) 0.5 M solution of NaPF_6 and 0.3 M solution of KPF_6 in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1 as electrolyte.

25

In an embodiment, the cathode active material of Formula (I) exhibits capacity fade ranging between 1.1-1.5 % over 250 cycles.

In an embodiment, the cathode active material of Formula (I) of the present invention find application in alkali ion-cell, in energy storage devices such as

30

batteries, rechargeable batteries, electrochemical devices and electrochromic devices.

In another embodiment, the present invention discloses the battery, rechargeable
5 battery comprising the sodium-ion cell or the electrochemical devices with said cathode active material of Formula (I) arranged in series, parallel, or both.

The following examples which include the preferred embodiments, will serve to illustrate the practice of this invention, it being understood that the particulars
10 shown are by way of example and for purpose of illustrative discussion of preferred embodiments of the invention.

Examples:

The advantages of the present invention are discussed with regard to experimental
15 examples.

Example 1: Composites of $\text{Na}_{0.657}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$

The composite $\text{Na}_{0.657}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$ is used as cathode active material, along with hard carbon as anode material. Further, 0.5M solution of NaPF_6 in a mixture
20 of ethylene carbonate, diethyl carbonate and Fluoroethylene carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1 is employed as electrolyte.

The cells were formed at 0.1C rate and post formation, were run at 0.5C rate. To ensure that the cell was fully charged, the cell was potentiostatically held at 4.2 V
25 at the end of the constant current charging process for 1 hour during the formation cycling. For the rest of the tests after the formation cycling, cells were only held for 1 minute potentiostatically at 4.2V to ensure full desaturation of the cathode. The testing was carried out at 25°C.

30 During the cell charging process, sodium ions are extracted from the cathode active material, and inserted into the Hard Carbon anode. During the subsequent

discharge process, sodium ions are extracted from the Hard Carbon and re-inserted into the cathode active material.

The constant current cycling graphs are plotted in **Figure 1(a)** and **Figure 1(b)**.

5

Figure 1 (a) illustrates the cell voltage profile. The graphical illustration in Figure 1(a) shows cell voltage (V) versus cathode specific capacity [mAh/g] for the first four charge/discharge cycles of the cell having $\text{Na}_{0.657}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$ used as cathode active material, and hard carbon as anode material.

10

Figure 1 (b) shows the constant current cycle life profile, i.e. the relationship between cathode specific capacity for discharge (mAh/g) and cycle number for hard carbon as anode material and $\text{Na}_{0.657}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$ as cathode material. For the first cycle, the discharge specific capacity for the cathode is about 80mAh/g.

15 For the twentieth cycle, the discharge specific capacity for the cathode is about 70 mAh/g. Therefore, it is evident that there occurs a capacity fade of about 12.5 % over 20 cycles, or an average of 0.625 % per cycle. The cathode material of this example demonstrates relatively poor capacity retention behavior.

20 **Example 2: Composites $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$ as cathode material**

In the present example, the cathode active material composite is selected as $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.571}\text{O}_2$ whereas the anode material is hard carbon. The electrolyte used is a 0.5M solution of NaPF_6 and 0.3M solution of KPF_6 in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1. The cells were formed at 0.1C rate and post formation, were run at 0.5C rate. To ensure that the cell was fully charged, the cell was potentiostatically held at 4.2 V at the end of the constant current charging process for 1 hour during the formation cycle. For the rest of the tests after the formation cycle, cells were only held for 1 minute potentiostatically at 4.2V to ensure full desaturation of the cathode. The testing was carried out at 25°C. During the cell charging process, host cations i.e. sodium and potassium are extracted

30

from the cathode active material and inserted into the anode made of hard carbon. During the subsequent discharge process, said host ions are extracted from the hard carbon and re-inserted into the cathode active material.

- 5 Figure 2 (A) shows the cell voltage profile, which is a graph of cell voltage (V) versus cathode specific capacity (mAh/g) for the first four charge/discharge cycles of the cathode active material composite and anode material of the present example.
- 10 Figure 2 (B) shows the constant current cycle life profile, which is a graphical relationship between cathode specific capacity for discharge (mAh/g) and cycle number for the cell having composite as mentioned herein. For cycle1, the discharge specific capacity for the cathode is about 85mAh/g. For cycle20, the discharge specific capacity for the cathode is about 80mAh/g. This represents a
- 15 capacity fade of about 5.88 % over 20 cycles or an average of 0.29% per cycle. The cathode material under test clearly demonstrates relatively better capacity retention behavior.

Example 3: Composite of $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.567}\text{Mg}_{0.006}\text{O}_2$ as cathode active material

- 20 In the present example, the composite of $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.567}\text{Mg}_{0.006}\text{O}_2$ is used as cathode active material, whereas hard carbon is used as anode material. The electrolyte used was a 0.5M solution of NaPF_6 and 0.3M solution of KPF_6 in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene
- 25 Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1. The cells were formed at 0.1C rate and post formation, were run at 0.5C rate. To ensure that the cell was fully charged, the cell was potentiostatically held at 4.2 V at the end of the constant current charging process for 1 hour during the formation cycle. For the rest of the tests after the formation cycle, cells were only held for 1 minute potentiostatically
- 30 at 4.2V to ensure full desaturation of the cathode. The testing was carried out at 25°C.

During the cell charging process, the host ions are extracted from the cathode active material and inserted into the hard carbon anode. During the subsequent discharge process, host ions are extracted from the hard carbon and re-inserted into the cathode active material.

Figure 3 (a) shows the cell voltage profile, which is a graphical representation of cell voltage (V) versus cathode specific capacity (mAh/g) for the first four charge/discharge cycles of the cell with the present composites.

10

Figure 3 (b) shows the constant current cycle life profile, which shows the relationship between cathode specific capacity for discharge (mAh/g) and cycle number for the cell having composites of the present example. For cycle 1, the discharge specific capacity for the cathode is about 86mAh/g. For cycle 20, the discharge specific capacity for the cathode is about 82mAh/g. This represents a capacity fade of about 4.65 % over 20 cycles or an average of 0.23% per cycle. It may be concluded that the cell having cathode active material of the present example demonstrates relatively better capacity retention behavior.

20 **Example 4: Composite of $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_2$ as cathode active material**

In the present example, $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_2$ is employed as cathode active material whereas hard carbon is employed as anode material. The electrolyte used was a 0.5M solution of NaPF_6 and 0.3M solution of KPF_6 in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1. The cells were formed at 0.1C rate and post formation, were run at 0.5C rate. To ensure that the cell was fully charged, the cell was potentiostatically held at 4.2 V at the end of the constant current charging process for 1 hour during the formation cycle. For the rest of the tests after the formation cycle, cells were only held for 1 minute potentiostatically at 4.2V to ensure full desaturation of the cathode. The testing was carried out at 25°C.

30

During the cell charging process, the host ions are extracted from the cathode active material, and inserted into the hard carbon anode. During the subsequent discharge process, the host ions were extracted from the hard carbon and re-
5 inserted into the cathode active material.

Figure 4 (a) shows the cell voltage profile, that is graphical representation of cell voltage (V) versus cathode specific capacity (mAh/g) for the first four charge/discharge cycles of the composites of the present example.

10

Figure 4 (b) shows the relationship between cathode specific capacity for discharge (mAh/g) and cycle number for the composites of the present example. For cycle 1, the discharge specific capacity for the cathode is about 90mAh/g. For cycle 20, the discharge specific capacity for the cathode is about 87mAh/g. This
15 represents a capacity fade of about 3.33 % over 20 cycles or an average of 0.16% per cycle. The aforesaid cathode active material exhibits better capacity retention behavior than the previous composites.

Example 5: Composites of $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_{1.99}\text{F}_{0.02}$ as active cathode material

20

The present example includes $\text{Na}_{0.651}\text{K}_{0.007}\text{Cu}_{0.314}\text{Mn}_{0.564}\text{Mg}_{0.006}\text{Ti}_{0.006}\text{O}_{1.99}\text{F}_{0.02}$ as active cathode material whereas hard carbon is anode material. The electrolyte used was a 0.5M solution of NaPF_6 and 0.3M solution of KPF_6 in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate
25 (EC:DEC:FEC) in a ratio of 1:1:0.1. The cells were formed at 0.1C rate and post formation, were run at 0.5C rate. To ensure that the cell was fully charged, the cell was potentiostatically held at 4.2 V at the end of the constant current charging process for 1 hour during the formation cycle. For the rest of the tests after the formation cycle, cells were only held for 1 minute potentiostatically at 4.2V to
30 ensure full desaturation of the cathode. The testing was carried out at 25°C.

During the cell charging process, the host ions were extracted from the cathode active material and inserted into the hard carbon anode. During the subsequent discharge process, the host ions were extracted from the hard carbon and re-inserted into the cathode active material.

5

Figure 5 (a) shows the cell Voltage (V) versus cathode specific capacity (mAh/g) for the first 4 charge/discharge cycles of the said composites.

Figure 5 (b) shows the constant current cycle life profile, i.e. the relationship between cathode specific capacity for discharge (mAh/g) and cycle number for the present composite. For cycle1, the discharge specific capacity for the cathode is about 89mAh/g. For cycle 20, the discharge specific capacity for the cathode is about 88 mAh/g. This represents a capacity fade of about 1.12 % over 250 cycles or an average of 0.0044% per cycle. Hence, the cathode active material of the present example shows excellent capacity retention behavior, which is better than the rest of exemplary composite.

Although the invention has been described in detail in the foregoing for the purpose of illustration, it is to be understood that such detail is solely for that purpose and that variations can be made therein by those skilled in the art without departing from the spirit and scope of the invention except as it may be limited by the claims.

25

We claim,

1. Mixed cation doped cuprate cathode active material, with little or no fading on cycling, of the formula (I) for energy devices comprising,

5



wherein

‘A’ comprises one or more alkali metals selected from lithium, sodium,
10 potassium and cesium;

‘B’ comprises one or more alkali metals selected from lithium, sodium,
potassium and cesium;

Cu is copper in oxidation state 2+,

Mn is manganese in oxidation state 4+,

15 E^1 comprises one or more elements in oxidation state 2+,

E^2 comprises one or more elements in oxidation state 4+, and

E^3 comprises one or more elements in oxidation state 1-

wherein

$0.4 \leq a \leq 0.9$, preferably $0.5 \leq a \leq 0.85$, further preferably $0.6 \leq a \leq 0.75$,

20 $0.001 \leq b \leq 0.4$, preferably $0.005 \leq b \leq 0.1$, further preferably $0.005 \leq b \leq 0.04$,

$0 \leq v \leq 0.5$, preferably $0 \leq v \leq 0.45$ and ideally $0 \leq v \leq 0.333$,

$0.4 \leq w \leq 0.8$, preferably $0.45 \leq w \leq 0.7$ and ideally $0.5 \leq w \leq 0.7$,

at least one of x and y is in the range of 0 to 0.39,

z is in the range of 0 to 1.99.

- 25 2. The mixed cation doped cuprate cathode active material as claimed in claim 1, comprising;

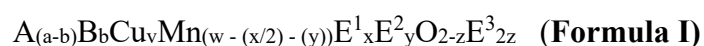
i. $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.571}O_2$,

ii. $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.567}Mg_{0.006}O_2$,

iii. $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.564}Mg_{0.006}Ti_{0.006}O_2$,

30 iv. $Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.564}Mg_{0.006}Ti_{0.006}O_{1.99}F_{0.02}$.

3. The mixed cation doped cuprate cathode active material as claimed in claim 1, wherein the formed cuprate is doped with larger alkali ions that results in enlarged interlayer spacing, providing larger channels for ion movement; the +2 and +4 doped elements reinforce the manganese back-bone and prohibits phase transitions and the degradation over multiple cycles; the anionic doping with a monovalent atom of 1- oxidation state increases the strength of the bonds and reduces oxygen loss at high potentials.
4. The mixed cation doped cuprate cathode active material as claimed in claim 1, wherein said cathode active material is in conjunction with a counter electrode and one or more electrolyte materials in energy storage devices.
5. The mixed cation doped cuprate cathode active material as claimed in claim 1, wherein a charge-discharge profile of the electrode exhibits two plateaus at about 4V and 3.6V in energy storage devices comprising hard carbon as anode and 0.5M solution of NaPF₆ and 0.3M solution of KPF₆ in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1 as electrolyte.
6. A mixed cation doped cuprate cathode active material comprising Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.564}Mg_{0.006}Ti_{0.006}O_{1.99}F_{0.02} with capacity fade ranging between 1.1 to 1.5 % over 250 cycles.
7. The alkali-ion electrochemical cell comprising mixed cation doped cuprate cathode active material of formula (I);



wherein

‘A’ comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

'B' comprises one or more alkali metals selected from lithium, sodium, potassium and cesium;

Cu is copper in oxidation state 2+,

Mn is manganese in oxidation state 4+,

5 E¹ comprises one or more elements in oxidation state 2+,

E² comprises one or more elements in oxidation state 4+, and

E³ comprises one or more elements in oxidation state 1-

wherein

$0.4 \leq a \leq 0.9$, preferably $0.5 \leq a \leq 0.85$, further preferably $0.6 \leq a \leq 0.75$,

10 $0.001 \leq b \leq 0.4$, preferably $0.005 \leq b \leq 0.1$, further preferably $0.005 \leq b \leq 0.04$,

$0 \leq v \leq 0.5$, preferably $0 \leq v \leq 0.45$ and ideally $0 \leq v \leq 0.333$,

$0.4 \leq w \leq 0.8$, preferably $0.45 \leq w \leq 0.7$ and ideally $0.5 \leq w \leq 0.7$,

at least one of x and y is in the range of 0 to 0.39,

z is in the range of 0 to 1.99.

15

8. The alkali-ion electrochemical cell as claimed in claim 7, comprising;

(i) Hard carbon as negative electrode anode;

(ii) The cation doped cuprate active material of Formula (I) as positive electrode cathode with little or no fading;

20 (iii) A separator between the positive electrode and negative electrode; and

(iv) Electrolyte containing 0.5M solution of NaPF₆ and 0.3M solution of KPF₆ in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1.

25

9. The alkali ion- electrochemical cell as claimed in claim 8, comprising;

(i) Na_{0.651}K_{0.007}Cu_{0.314}Mn_{0.564}Mg_{0.006}Ti_{0.006}O_{1.99}F_{0.02} as cathode active material;

(ii) Hard Carbon as anode material;

30 (iii) A separator between the positive electrode and negative electrode; and

(iv) 0.5 M solution of NaPF₆ and 0.3 M solution of KPF₆ in a mixture of Ethylene Carbonate, Diethyl Carbonate and Fluoroethylene Carbonate (EC:DEC:FEC) in a ratio of 1:1:0.1 as electrolyte.

5 10. The alkali-ion electrochemical cell as claimed in claim 9, wherein said cell exhibits capacity fade ranging between 1.1-1.5 % over 250 cycles.

11. A battery comprising a plurality of electrochemical cells as claimed in claims 7 to 9 arranged in series, parallel, or both.

10

12. The mixed cation doped cuprate cathode active material cathode active material of Formula (I) as claimed in claim 1 for use in alkali ion-cell, in energy storage devices such as batteries, rechargeable batteries, electrochemical devices and electrochromic devices.

15

13. A process for synthesis of mixed cation doped cuprate cathode active material of formula (I) claimed in any one of the preceding claim comprising;

- (i) Intimately mixing together the starting materials (i.e. the precursors for the cathode active material) in the correct stoichiometric ratio and pressing into a pellet;
- 20 (ii) Heating the resulting mixture in a furnace under a suitable atmosphere comprising ambient air, or an inert atmosphere of argon or nitrogen, wherein the gases may be flowing if desired, at a single temperature or over a range of temperatures between
- 25 450°C and 800°C until reaction product forms; and optionally
- (iii) Allowing the product to cool before grinding it to a powder.

Dated this 10th day of April, 2021

30


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MIXED CATION DOPED CUPRATE CATHODES AND PROCESS THEREOF

ABSTRACT:

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The present invention discloses mixed cation doped cuprate cathodes for energy devices notably non-aqueous alkali-ion rechargeable electrochemical cells and batteries and to the process of preparation thereof. More particularly, the present invention relates to doped cuprate cathode active material of Formula (I) that shows little or no fading on cycling, which is able to be recharged multiple times without significant loss in charge capacity.

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Applicant:	Sheet No.: 1

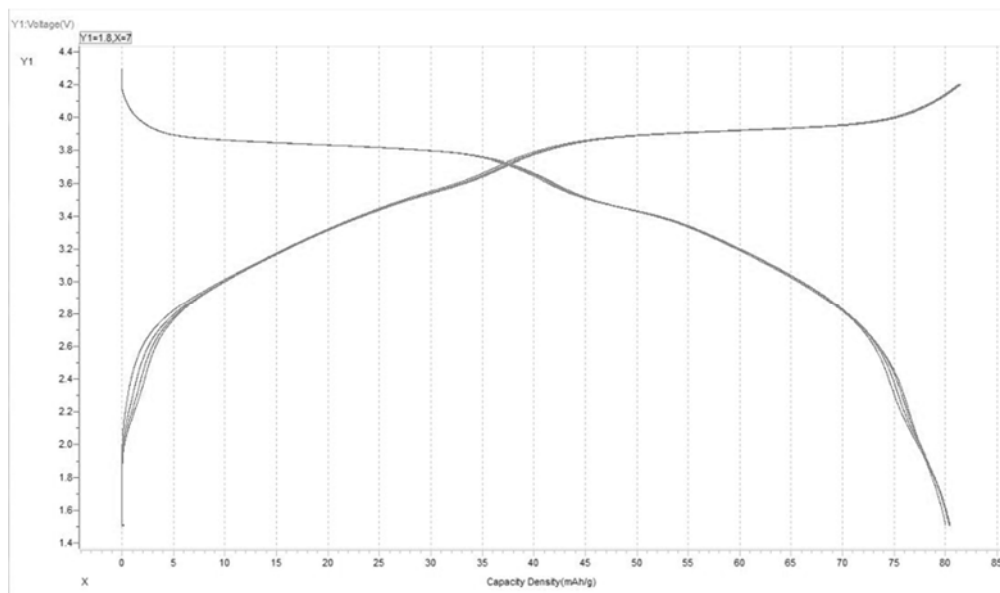


Fig 1 (a)

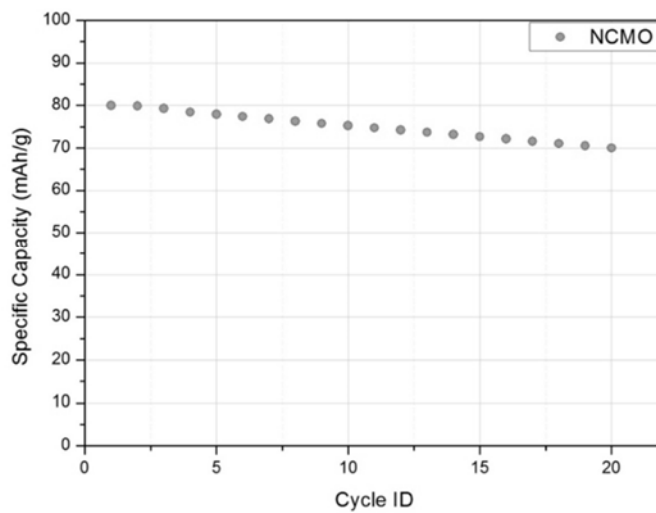


Fig 1(b)

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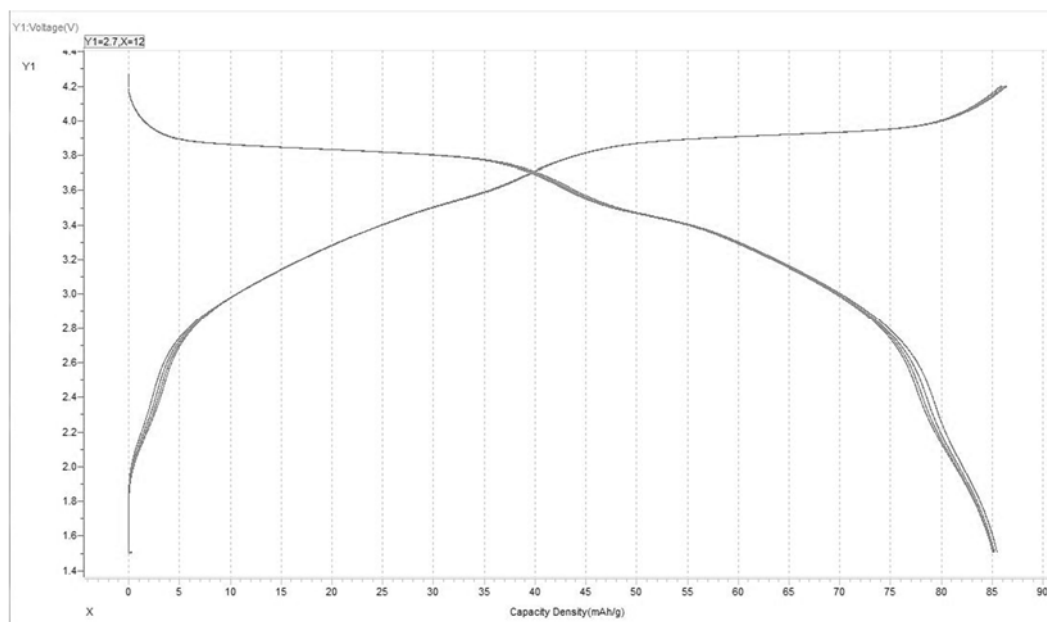


Fig 2(a)

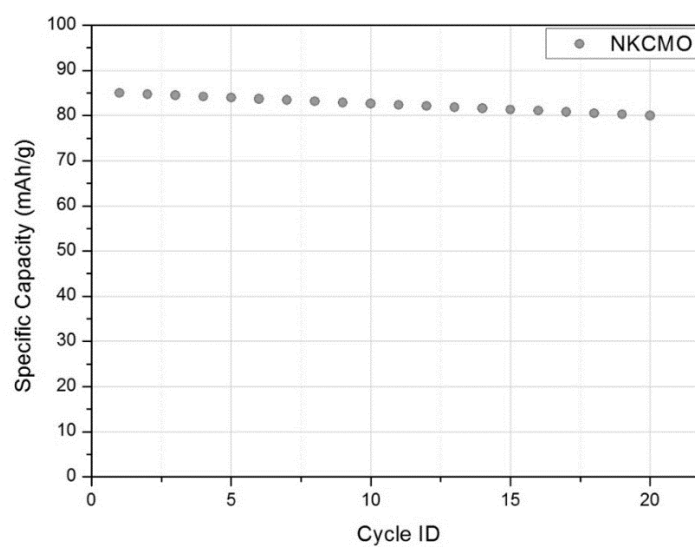


Fig 2(b)

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Applicant:	Sheet No.: 3

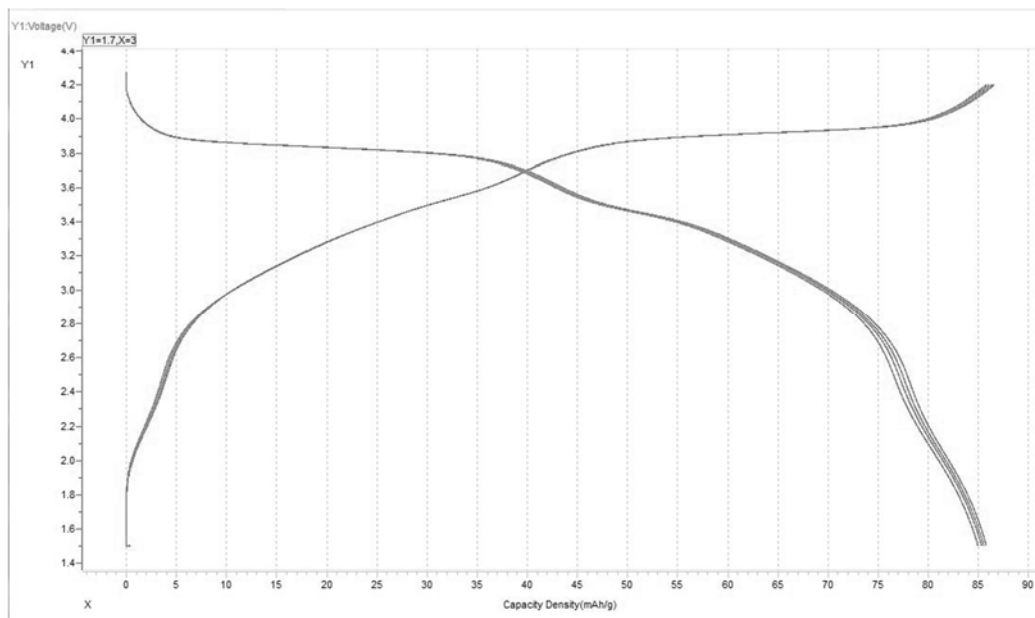


Fig 3(a)

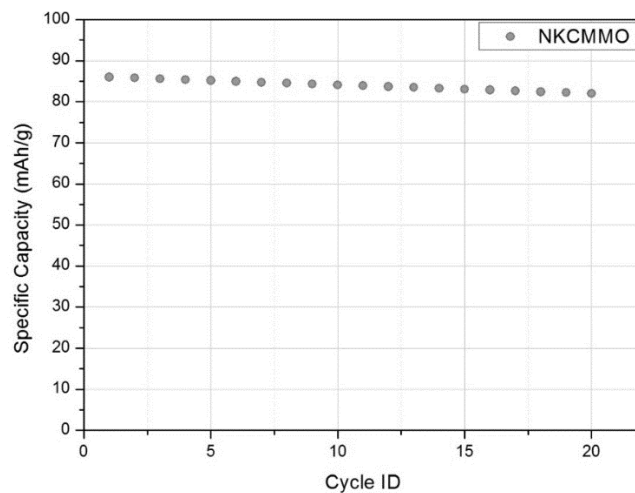


Fig 3(b)

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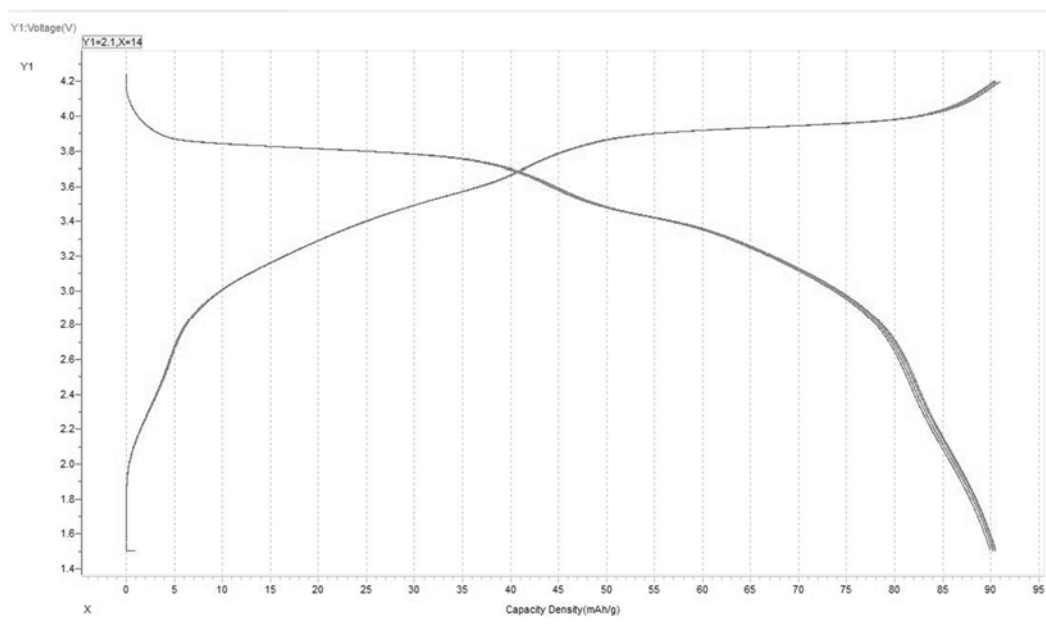


Fig 4(a)

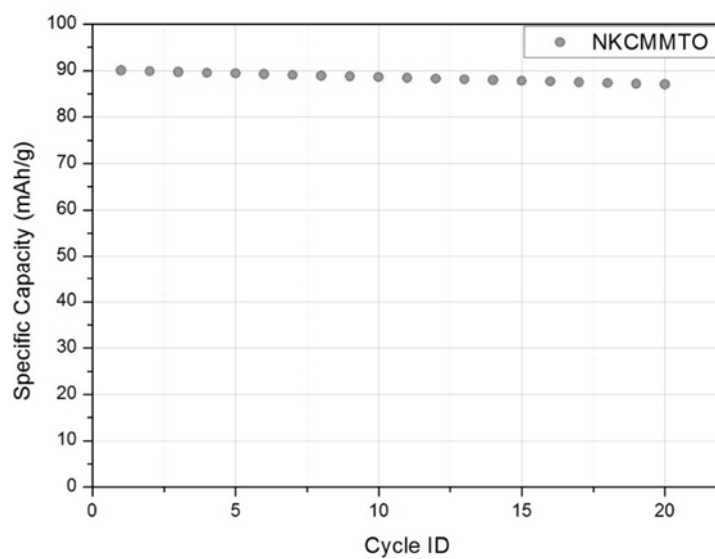


Fig 4(b)

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Applicant:	Sheet No.: 5

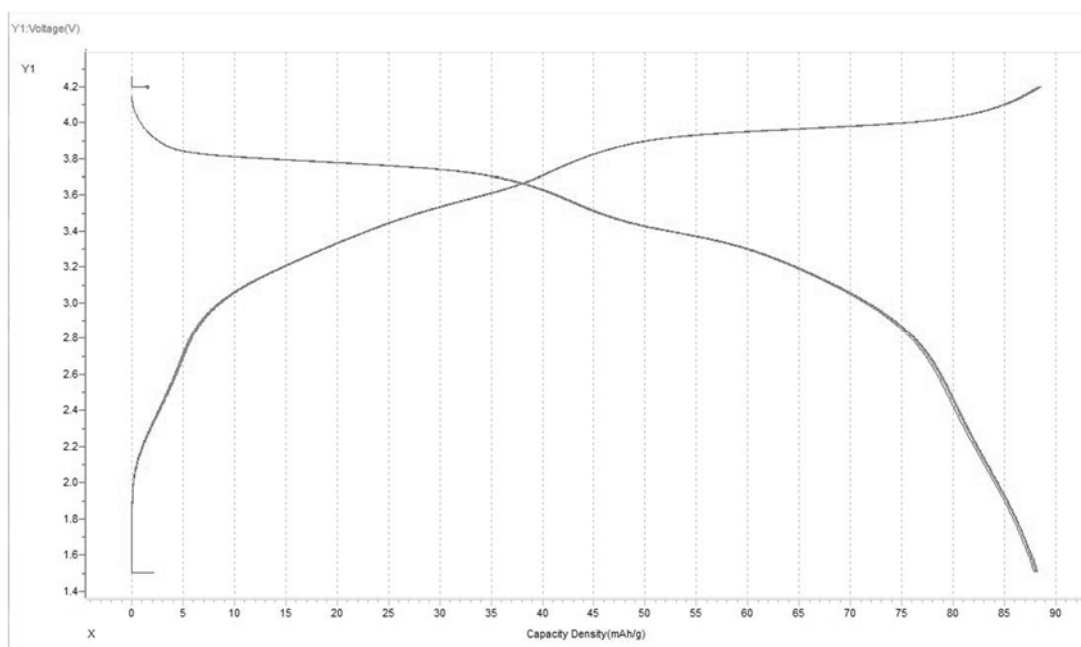


Fig 5(a)

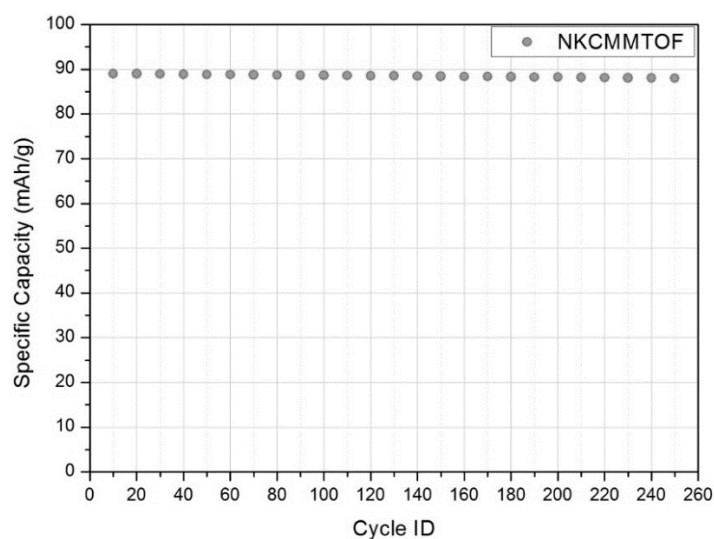


Fig 5(b)

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