

A Novel Cathodic Combination in Sodium-Ion Battery Based on $\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, Na_2MnO_3 , and NaCoO_2 Combination: Synthesis and Characterization

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Abstract: Several combinations, including $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, $x\text{Na}_2\text{MnO}_3$, and $y\text{NaCoO}_2$ systems, have been prepared by the sol-gel methods. Stoichiometric amounts of the NaNO_3 , $\text{Mn}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$, $\text{Co}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$, $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ for testing of 28 samples have been applied. We demonstrated that samples, including $\text{Na}_{1.5}\text{Ni}_{0.117}\text{Co}_{0.366}\text{Al}_{0.017}\text{Mn}_{0.5}\text{O}_2$ with Al-doped are the best composition for cathode materials in NIBTs due to the amount of cobalt (Co) in this combination, that is lower compared to NiCoO_2 . This is important in the viewpoint of expenditure and toxic position of Co usage. Charges and discharges behavior of the cathode materials were analyzed via performing cycle tests in the range of 2.0-4.0V. Our results have shown that these kinds of samples could help to remove the unsuitable disadvantage of Co with high efficiency and also to replace Na that is cheaper instead of Li.

Keywords: sodium ion battery; NaCoO_2 ; Na_2MnO_3 ; sol-gel method.

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1. Introduction

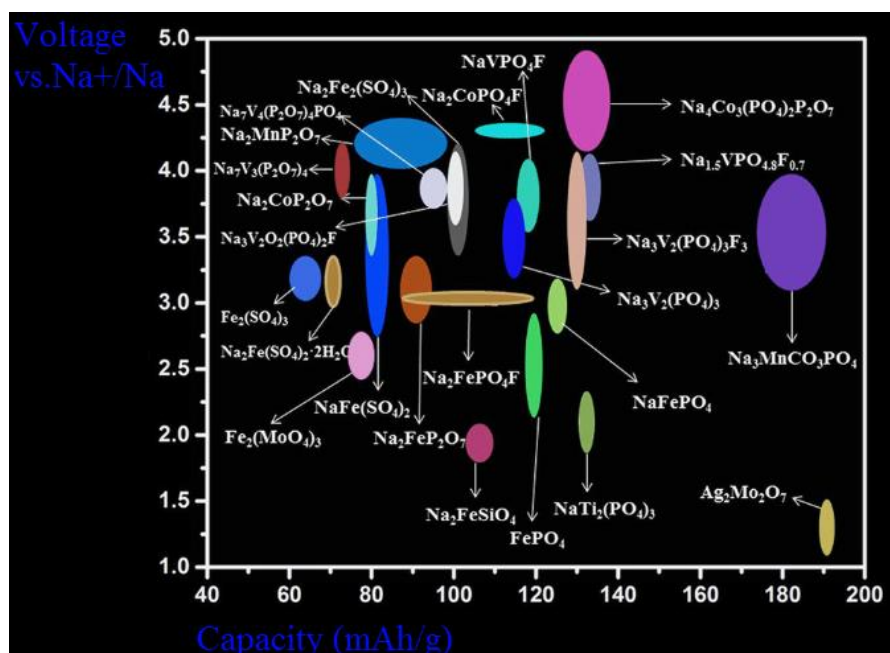
The cathode material elements such as manganese, nickel-cobalt (NMC), are important metals for sodium-ion batteries (NIBs) because of their abilities to operate in high voltages and large energies that can be stored in a small space. Although Ni elements give several high capacities for storing energies, they are unstable with the related electrolyte. In table 1, there are some advantages and some disadvantages items for the NMC of NIBs [1-5] comparing to NIBs. A major disadvantage of Co is its cost and toxicity, and generally, it is almost two times more costly to manufacture than the Ni. This is an important item due to any extra cost that is a basic issue. The safety factor is also a matter of crucial importance to the Na batteries that this problem appears in large size batteries as compared to the small size batteries, i.e., batteries for electric cars would have more risk of catching fire as compared to the cell phones. If a mixed solid solution is used, the explosion of batteries can be controlled, especially with doping of other elements such as Al. In addition, $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ doped with Al is suitable for the environment as compared to isolated cathode, such as NaCoO_2 [6-10]. The aim of this study is to find a solid ternary solution of Na-based cathode material in batteries replacement for NaCoO_2 , which is expensive and toxic. Therefore a combination of ternary composition diagrams including $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, $x\text{Na}_2\text{MnO}_3$, and $y\text{NaCoO}_2$ have been investigated. NIBs were the most interesting subject among the scientist for replacing NaCoO_2 (reported by Rossen *et al.*). In 1992 for the $\text{NaNi}_{0.5}\text{Mn}_{0.5}\text{O}_2$ [7-11]. A paper was reported in 1998 about

the electrochemical behavior that indicates Ni and Mn ions were present in “+2” and “+4” oxidation rather than the “+3”. They show at a high temperature, there is a large falling capacity after 25 cycles between 100 to 155 “mAhg⁻¹”, after 25 cycles, and to 75 “mAhg⁻¹” after 60 cycles. Nickel, which takes place between +2 and +4 valence positions, meanwhile manganese is +4, which remains without Jahn-Teller distortion with the +3 valence (Mn⁺³)[12-15].

Table 1. Commercial comparison between Lithium-ion battery and Sodium-ion battery.

| Commercial data | Lead-acid battery | LIBs | NIBs |
|-------------------|-----------------------------------------------|-----------------------------------------------------|---------------------------------------------|
| Cost | Low | High | Low |
| Energy Density | Low | High | Moderate/High |
| Safety | Moderate | Low | High |
| Materials | Toxic | Scarce | Earth-abundant |
| Cycling Stability | Moderate (high self-discharge) | High (negligible self-discharge) | High (negligible self-discharge) |
| Efficiency | Low (< 75%) | High (> 90%) | High (> 90%) |
| Temperature Range | -40 °C to 60 °C | -25 °C to 40 °C | -40 °C to 60 °C |
| Remarks | Mature technology; fast charging not possible | Transportation restrictions at the discharged state | Less mature technology; easy transportation |

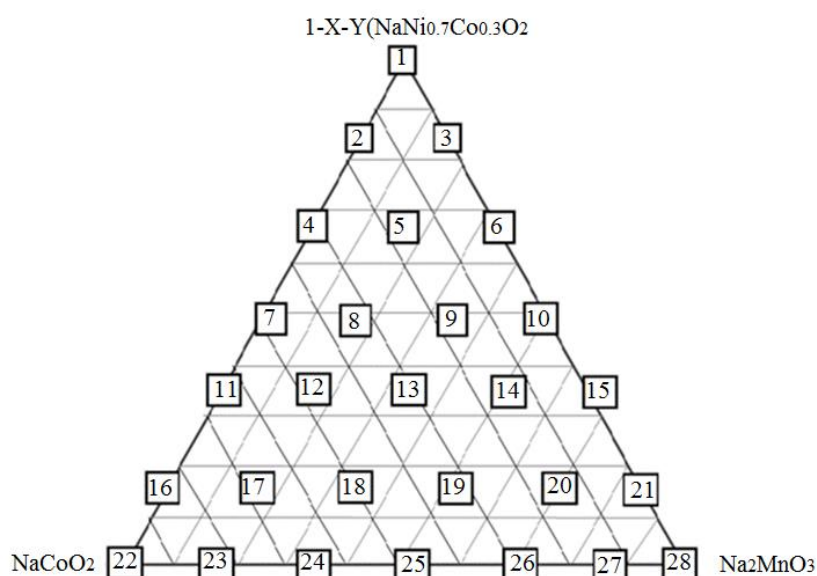
Moreover, mixing proper of various transition elements yield some advantageous efficiency of each led to the discovery of those kinds of materials. Therefore these materials are derived from Na (Ni_(1-x-y)Mn_xCo_y)O₂ that was published in 1999. They exhibited that the extra cobalt concentration would stabilize the composition blocks the Ni from entering the sodium layers. Meanwhile, Ni and Mn would provide structural stabilities and high capacities, respectively.



Scheme 1. Voltage-capacity diagram of various cathode materials in sodium-ion batteries

Too much of Co decrease capacities, and a large amount of Ni and Mn make problems in spinel structures; therefore, mixing of these elements must be optimized with suitable mole fraction. The layered compound of NaNi_{1/3}Mn_{1/3}Co_{1/3}O₂ can be synthesized and exhibited that this cathode has about 205 mAhg⁻¹ reversible discharge capacities in the range of 2.1-4.7 V, including large rate capabilities and thermal stabilities [16-21]. Scheme1. Voltage-Capacity curve of several cathode materials in NIB batteries for the appropriate stoichiometry of Na (Ni_xCo_(1-2x)Mn_x)O₂, x = 1/3, the medium oxidation numbers must be +3 same as Mn⁺⁴, Co⁺³, and Ni⁺², with the voltages in the ranges of, 2.1 up to 4.7 versus lithium ions concentration.

Moreover, about further electrochemical concepts, some extra works of $\text{NaNiO}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.2}\text{O}_2$, $\text{Na}(\text{Ni}_{0.8}\text{Co}_{0.2})\text{O}_2$, and $\text{NaNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$ cathodes have been accomplished (scheme 1). Although the rate capabilities of this material are smaller than that of NaCoO_2 , thermal stabilities are much better [32-36]. By this work, we look at the several compositions of binary and ternary solid solutions containing Na_2MnO_3 , NaCoO_2 , and $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2$ doped with Al (Scheme 2). Nowadays, Na_2MnO_3 has been selected for its suitable capacities, better safety, without any toxic and especially inexpensive as materials compared to NaCoO_2 [22-26].



Scheme 2. Binary and Ternary diagram of $\{(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2\}$, $x\text{Na}_2\text{MnO}_3$ and $y\text{NaCoO}_2$ compositions.

Binary and Ternary curve of $\{(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2\}$, $x\text{Na}_2\text{MnO}_3$, and $y\text{NaCoO}_2$ combinations, that can be denoted as $\text{Na}[\text{Na}_{1/3}\text{Mn}_{2/3}]\text{O}_2$, have the same structure as NaCoO_2 , except there is a super-lattice ordering of Na^+ and Mn^{4+} in transition metal shells. Na_2MnO_3 has a rock-like layer, including Na and Mn cations in alternating (1:2 ratio, respectively) situations, and these layers are divided through a cubic O_2 layer. As Mn is shown in +4 oxidation amount, it is impossible to be further oxidization in low voltages; so Na_2MnO_3 might be electrochemically inactive, whereas these materials in higher voltages, i.e., $>4.7\text{ V}$, will extract the Na cation through the behind of MnO_2 shells. Meanwhile, the mechanism is unilateral and not reversible so, only one atom of Na^+ ion can return during discharging towards the NaMnO_2 molecule. The extra Na atom that refracts within charging attaches to the higher capacities. Using of cathodes with Na_2MnO_3 is important due to simplifying the intercalation mechanism and also for providing structural stabilities during this intercalation [27-32]. Solid solutions were generally selected in terms of obtaining a cathode material with the superior capability of cycle-ability, capacities, and structural stabilities with lower costs (scheme 2).

By this work, the solid solutions have been used as a cathode material, including both binary and ternary systems. Solid solutions containing Na_2MnO_3 were found suitable for two major subjects, one the extraction of two Na^+ ions at voltages $>4.7\text{ V}$, where yields the extra amount of initial charges capacities, and second the MnO_2 ingredient that on the removal of Na, gives well structural stability in that combination. It is mentioned that the initial structures of the cathode material not only specified the initial characteristic capacity but also imply to the stability. The consistency of Na- dissociation cathode systems is substantial for any increase

of the cycle life. During the Na cation extracting, Ni and Co will be oxidized from +3 to +4 valance positions, so the structure cell volume will be changed. Al substitution in Co crystal is a suitable doped metal to increase stability and electrochemical efficiency. Al cation remains in the +3 valance position and has no contribution to its capacity only helps to reduce the structure volume changing and keeping the crystal stronger after a few cycling [33-39].

2. Materials and Methods

2.1. Experimental section.

The combinations were synthesized with the sol-gel method due to a simple chemical reaction, including low temperature and high homogeneity. Stoichiometric amounts of the NaNO_3 , $\text{Mn}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$, $\text{Co}(\text{Ac})_2 \cdot 4\text{H}_2\text{O}$, $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ as starting materials of sodium, manganese, cobalt, and nickel, in 28 samples of $\{[(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2]$ doped with Al, have been applied for several experimental testing. These mixtures were first dissolved in 50ml of DI H_2O , and then equivalence-molar weights of citric acid were added. The whole mixtures were heated via a water bath at 95°C and during the heating process, a clear, pink solution formed. Finally, the clear solution was slowly dried and turned into gel. This solution was continuously stirred for about 30 mins for the formation of a homogeneous mixture of gel and is also kept under a hot-plate for 13-15 hrs around 95°C . It is notable; acetate requires low and high heating temperatures to form a proper phase. The beaker containing the gel is then kept in a furnace and is heated to 460°C for 4-6 hrs. Usually, what occurs during this stage is that whole of the acetates present would get burned off, but the desired phases are not formed. The heat-treated products were ground in an agate mortar to obtain powders, and then the powder was calcined at $850\text{-}900^\circ\text{C}$ for 11-13 hrs. The second heating is done at higher temperatures due to the wide range of materials present in the combination and also, more importantly, to ensure the formation of a correct phase for the crystalline structure then the material collected from the furnace is stored at a dry place until it is made into a cathode. The prepared products were first mixed with acetylene black and NMP. Therefore in the system of $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.2}\text{Al}_{0.1}$, $x\text{Na}_2\text{MnO}_3$, $y\text{NaCoO}_2$ (table 2), the high Ni content usually gives higher initial specific capacities while Co and Al increase the stabilities and life cycles.

3. Results and Discussion

This work (based on our previous works [40-97]) is used for explaining the best cathode material, including $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, $x\text{Na}_2\text{MnO}_3$, $y\text{NaCoO}_2$ systems with high initial discharge capacity, grate cyclability, and inexpensive compared to usual NIBs cathode materials. Therefore, 28 different structures using the lever rule, stoichiometric weights, and mole-fractions were selected in order to find an optimized material with suitable efficiency. These 28 points were selected via the triangle phase diagram (scheme 2 and table 2&3) and also synthesis through the sol-gel method. Ni and Al amount are decreased towards the down direction of the triangle; meanwhile, the compositions of 22, 24 have zero Ni and Al percentage. A high Mn value is found in sample 22, and its content decreases at the opposite endpoints of the triangle. Co percentage is appeared in a wide region in the triangle and also decreases near $\text{Na}(\text{Na}_{0.33}\text{Mn}_{0.66})\text{O}_2$. It is predicted which capacities and cyclability of these systems are directly related to the amount of Mn, Co, Ni, and Al. Therefore, specific capacity is determined as the number of energies which can be reserve in volume or mass (Ah), while the rate capability can be determined as the rate at which the cell is being charged.

Table 2. 28 different composition points according to using the lever rule, stoichiometric weights, and mole fractions of the triangle diagram

| Sample | Composition | Al-doped |
|--------|------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------|
| 1 | NaNi _{0.7} Co _{0.3} O ₂ | NaNi _{0.7} Co _{0.2} Al _{0.1} O ₂ |
| 2 | Na _{1.167} Ni _{0.583} Co _{0.25} Mn _{0.167} O ₂ | Na _{1.167} Ni _{0.583} Co _{0.167} Al _{0.083} Mn _{0.167} O ₂ |
| 3 | NaNi _{0.583} Co _{0.417} O ₂ | NaNi _{0.583} Co _{0.334} Al _{0.083} O ₂ |
| 4 | Na _{1.333} Ni _{0.467} Co _{0.203} Mn _{0.333} O ₂ | Na _{1.333} Ni _{0.467} Co _{0.133} Al _{0.067} Mn _{0.333} O ₂ |
| 5 | Na _{1.167} Ni _{0.467} Co _{0.366} Mn _{0.167} O ₂ | Na _{1.167} Ni _{0.467} Co _{0.299} Al _{0.067} Mn _{0.167} O ₂ |
| 6 | NaNi _{0.467} Co _{0.533} O ₂ | NaNi _{0.467} Co _{0.466} Al _{0.067} O ₂ |
| 7 | Na _{1.5} Ni _{0.35} Co _{0.15} Mn _{0.5} O ₂ | Na _{1.5} Ni _{0.35} Co _{0.1} Al _{0.05} Mn _{0.5} O ₂ |
| 8 | Na _{1.333} Ni _{0.35} Co _{0.317} Mn _{0.333} O ₂ | Na _{1.333} Ni _{0.35} Co _{0.267} Al _{0.05} Mn _{0.333} O ₂ |
| 9 | Na _{1.167} Ni _{0.35} Co _{0.483} Mn _{0.167} O ₂ | Na _{1.167} Ni _{0.35} Co _{0.433} Al _{0.05} Mn _{0.167} O ₂ |
| 10 | NaNi _{0.35} Co _{0.6} O ₂ | NaNi _{0.35} Co _{0.6} Al _{0.05} O ₂ |
| 11 | Na _{1.667} Ni _{0.233} Co _{0.1} Mn _{0.667} O ₂ | Na _{1.667} Ni _{0.233} Co _{0.067} Al _{0.033} Mn _{0.667} O ₂ |
| 12 | Na _{1.5} Ni _{0.233} Co _{0.267} Mn _{0.5} O ₂ | Na _{1.5} Ni _{0.233} Co _{0.234} Al _{0.033} Mn _{0.5} O ₂ |
| 13 | Na _{1.333} Ni _{0.233} Co _{0.434} Mn _{0.333} O ₂ | Na _{1.333} Ni _{0.233} Co _{0.4} Al _{0.033} Mn _{0.333} O ₂ |
| 14 | Na _{1.167} Ni _{0.233} Co _{0.6} Mn _{0.167} O ₂ | Na _{0.167} Ni _{0.233} Co _{0.567} Al _{0.033} Mn _{0.167} O ₂ |
| 15 | NaNi _{0.233} Co _{0.767} O ₂ | NaNi _{0.233} Co _{0.734} Al _{0.033} O ₂ |
| 16 | Na _{1.833} Ni _{0.116} Co _{0.049} Mn _{0.833} O ₂ | Na _{1.833} Ni _{0.117} Co _{0.033} Al _{0.017} Mn _{0.833} O ₂ |
| 17 | Na _{1.667} Ni _{0.116} Co _{0.217} Mn _{0.667} O ₂ | Na _{1.667} Ni _{0.117} Co _{0.199} Al _{0.017} Mn _{0.667} O ₂ |
| 18 | Na _{1.5} Ni _{0.116} Co _{0.384} Mn _{0.5} O ₂ | Na _{1.5} Ni _{0.117} Co _{0.366} Al _{0.017} Mn _{0.5} O ₂ |
| 19 | Na _{1.333} Ni _{0.116} Co _{0.551} Mn _{0.333} O ₂ | Na _{1.333} Ni _{0.117} Co _{0.533} Al _{0.017} Mn _{0.333} O ₂ |
| 20 | Na _{1.167} Ni _{0.116} Co _{0.717} Mn _{0.167} O ₂ | Na _{1.167} Ni _{0.117} Co _{0.699} Al _{0.017} Mn _{0.167} O ₂ |
| 21 | NaNi _{0.116} Co _{0.884} O ₂ | NaNi _{0.117} Co _{0.866} Al _{0.017} O ₂ |
| 22 | Na ₂ MnO ₂ | Na ₂ MnO ₂ |
| 23 | Na _{1.833} Co _{0.167} Mn _{0.833} O ₂ | Na _{1.833} Co _{0.167} Mn _{0.833} O ₂ |
| 24 | Na _{1.667} Co _{0.333} Mn _{0.667} O ₂ | Na _{1.667} Co _{0.333} Mn _{0.667} O ₂ |
| 25 | Na _{1.5} Co _{0.5} Mn _{0.5} O ₂ | Na _{1.5} Co _{0.5} Mn _{0.5} O ₂ |
| 26 | Na _{1.333} Co _{0.667} Mn _{0.333} O ₂ | Na _{1.333} Co _{0.667} Mn _{0.333} O ₂ |
| 27 | Na _{1.167} Co _{0.833} Mn _{0.167} O ₂ | Na _{1.167} Co _{0.833} Mn _{0.167} O ₂ |
| 28 | NaCoO ₂ | NaCoO ₂ |

Obviously, the C-rate is the capacity of the battery separated by the hourly charging rate. The discharge capacity diagrams have been compared with the 18650-type “C/NaCo₂ Sony battery” result, which is designed by Ehrlich. Since NaCoO₂ cathode has an initial discharge capacity around 140 mAhg⁻¹, therefore any materials of these combinations with initial capacities more than this amount can be selected. The samples were tested via a cyclor Arbin BT 2000 tester, in the range of 2.2 V to the 4.8 V, with a low C-rate of C/12. The initial results indicate a wide range and irregular cyclability and capacities and consequently, the initial discharge capacities varied between 100 mAhg⁻¹ to 250 “mAhg⁻¹”. Both capacity and cyclability increase from NaNi_{0.7}Co_{0.3}Al_{0.1}O₂ towards the binary composition of Na₂MnO₃ and NaCoO₂.

Table 3. Summary capacity and cyclability for 28 samples of the system [(1-x-y)NaNi_{0.7}Co_{0.2}Al_{0.1}]O₂.

| Sample | Blend | Na ₂ MnO ₃ | NaCoO ₂ | NaNi _{0.7} Co _{0.2} Al _{0.1} O ₂ | Capacity | Cyclability |
|--------|---------|----------------------------------|--------------------|------------------------------------------------------------------------|----------|-------------|
| 1 | Pure | 0 | 0 | 1 | 80.5 | 61 |
| 2 | Binary | 0.167 | 0 | 0.833 | 92.3 | 66 |
| 3 | Binary | 0 | 0.167 | 0.833 | 99.2 | 85 |
| 4 | Binary | 0.333 | 0 | 0.667 | 105.5 | 62 |
| 5 | Ternary | 0.167 | 0.167 | 0.667 | 130.5 | 60 |
| 6 | Binary | 0 | 0.333 | 0.667 | 88.5 | 85 |
| 7 | Binary | 0.500 | 0 | 0.500 | 115.5 | 81 |
| 8 | Ternary | 0.333 | 0.167 | 0.500 | 107.2 | 61 |
| 9 | Ternary | 0.167 | 0.333 | 0.500 | 113.1 | 81 |
| 10 | Binary | 0 | 0.500 | 0.500 | 119.3 | 79 |
| 11 | Binary | 0.667 | 0 | 0.333 | 124.7 | 99 |
| 12 | Ternary | 0.500 | 0.167 | 0.333 | 113.1 | 90 |
| 13 | Ternary | 0.333 | 0.333 | 0.333 | 170.1 | 88 |
| 14 | Ternary | 0.167 | 0.500 | 0.333 | 163.5 | 85 |
| 15 | Binary | 0 | 0.667 | 0.333 | 120.6 | 95 |
| 16 | Binary | 0.833 | 0 | 0.167 | 171.5 | 65 |

| Sample | Blend | Na ₂ MnO ₃ | NaCoO ₂ | NaNi _{0.7} Co _{0.2} Al _{0.1} O ₂ | Capacity | Cyclability |
|--------|---------|----------------------------------|--------------------|------------------------------------------------------------------------|----------|-------------|
| 17 | Ternary | 0.667 | 0.167 | 0.167 | 131.5 | 91 |
| 18 | Ternary | 0.500 | 0.333 | 0.167 | 210.2* | 99* |
| 19 | Ternary | 0.333 | 0.5 | 0.167 | 119.3 | 92 |
| 20 | Ternary | 0.167 | 0.667 | 0.167 | 218.1 | 54 |
| 21 | Binary | 0 | 0.833 | 0.167 | 179.5 | 92 |
| 22 | Pure | 1 | 0 | 0 | 71.5 | 90 |
| 23 | Binary | 0.833 | 0.167 | 0 | 103.5 | 89 |
| 24 | Binary | 0.667 | 0.333 | 0 | 115.2 | 90 |
| 25 | Binary | 0.500 | 0.500 | 0 | 203.2 | 90 |
| 26 | Binary | 0.333 | 0.667 | 0 | 179.5 | 46 |
| 27 | Binary | 0.167 | 0.833 | 0 | 215.5 | 75 |
| 28 | Pure | 0 | 1 | 0 | 230.2 | 85 |

Although sample 20 shows a high capacity of “248.1” mAhg⁻¹, it contains a low cyclability. Sample 25 and 18 exhibit a good capacity of 201.2 and 220.2 mAhg⁻¹, respectively, with high cyclability; meanwhile because of Mn⁴⁺ ion, sample 22 has a low capacity.

Ternary Graph of Y = Capacity versus X₁, X₂, X₃
 $Y=150X_1+220X_2 + 90X_3$

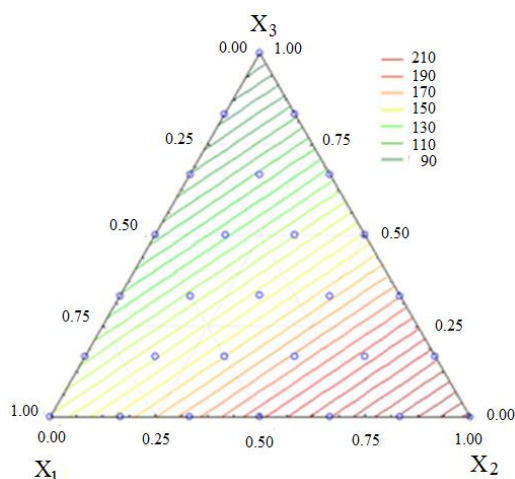


Figure 1. Ternary of capacity versus variable (1), variable (2) and variable (3) for 28 samples of (1-x-y)NaNi_{0.7}Co_{0.2}Al_{0.1}, xNa₂MnO₃, yNaCoO₂ composites.

Although this information is not enough for determining a suitable cathode material, the statistical analysis can be applied for explaining the region of the best item from the data of the 28 compositions. Therefore any testing with both capacity and cyclability relation in the viewpoint of the triangle regions is needed.

Ternary Graph of Y = Capacity versus X₁, X₂, X₃
 $Y= 88X_1+88X_2-21X_3$

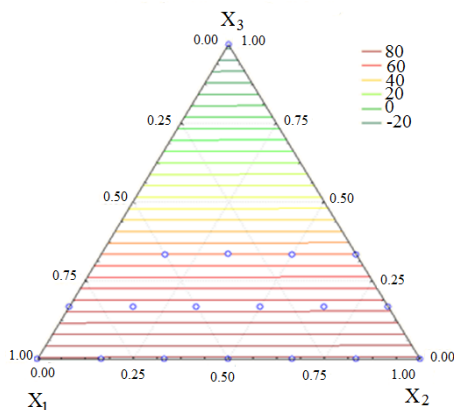


Figure 2. Ternary of cyclability versus X₁, X₂, and X₃ for 28 samples of (1-x-y)NaNi_{0.7}Co_{0.2}Al_{0.1}, xNa₂MnO₃, yNaCoO₂ composites.

In this work, the “SATISTICA” software has been selected for analyzing the data of variable (1), variable (2), and variable (3), which are Na_2MnO_3 , NaCoO_2 , and $\text{NaNi}_{0.7}\text{Co}_{0.2}\text{Al}_{0.1}\text{O}_2$ mole-fraction respectively and also the variable (4) which are capacity and cyclability alternately (table 3 and figs 1&2).

By this work, several compositions are used to explore the novel cathode materials with two-dimensional layered form through the ternary composition’s diagram among Iso-structural material. Although Na_2MnO_3 or $\text{Na}[\text{Na}_{1/3}\text{Mn}_{2/3}]\text{O}_2$ has a structure similar to NaCoO_2 , there is the super-lattice ordering of Mn^{4+} and Na^+ in transition layers. $\text{NaNi}_{0.7}\text{Co}_{0.2}\text{Al}_{0.1}\text{O}_2$ can be demonstrated as a solid solution between NaCoO_2 and NaNiO_2 with Al-doped in Co site, which is a promising cathode material due to its improved stabilities and electrochemical performances.

Ternary Graph of Y = Capacity versus X_1, X_2, X_3

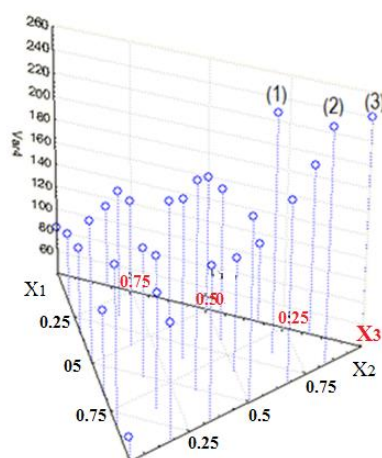


Figure 3. Three samples of $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.2}\text{Al}_{0.1}\text{O}_2 = X_3$, $x\text{Na}_2\text{MnO}_3 = X_1$, $y\text{NaCoO}_2 = X_2$ composites with high precisionist of capacity and cyclability versus X_1, X_2 , and X_3 .

With more testing, three samples were chosen and subsequently synthesized, characterized, and tested consequently with high precisionists. Via initial discharge capacity and some other information, numbers 18, 27, and 28 are indicated as the best cathode material among those structures (figs. 3&4, table 2&3). As the number 28 is pure material (NaCoO_2), and number 27 is binary, the number 18 “ $\text{Na}_{1.5}\text{Ni}_{0.117}\text{Co}_{0.366}\text{Al}_{0.017}\text{Mn}_{0.5}\text{O}_2$ ” is suggested as the best combination for cathode material.

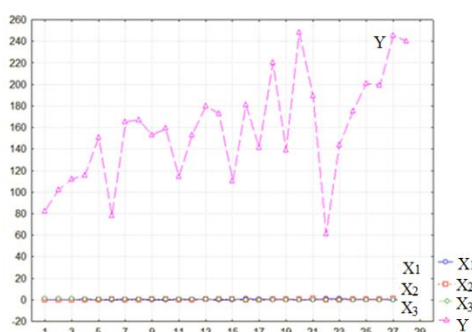


Figure 4. Multiple variable combinations of capacities and cyclability are based on the mole-fraction of X_1, X_2 , and X_3 in 28 compositions.

The repeat of sample 20 was made into T-Cells and subjected to electrochemical testing using the original conditions, and the cycling was done between 2.1-4.7 V with constant C-rate of C/12 at room temperature (fig. 5). The repeated sample shows suitable charge and discharge capacities (discharge capacity of $150, \text{mAhg}^{-1}$) than the original sample. With the same method,

we used $(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2$, $x\text{Na}_2\text{MnO}_3$, and $y\text{NaCoO}_2$ composites; the sample $\text{Na}_{1.5}\text{Ni}_{0.117}\text{Co}_{0.366}\text{Al}_{0.017}\text{Mn}_{0.5}\text{O}_2$ has been tested for comparing in viewpoints of capacity and cyclability (Fig. 5), the repeat sample exhibits in the same mole fraction, the system with Al-doped has a better charge, and discharge capacities (discharge capacity of $155, \text{mAhg}^{-1}$) compared to the non-Al-doped system (discharge capacity of $144, \text{mAhg}^{-1}$).

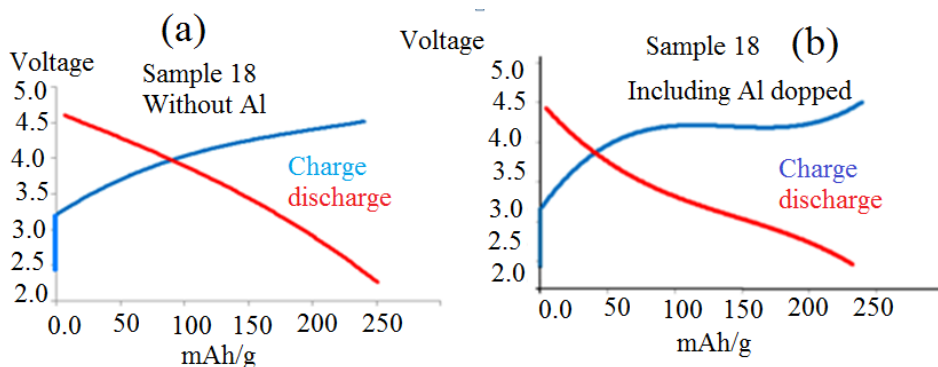


Figure 5. Charge and discharge capacity of (a): $\text{Na}_{1.5}\text{Ni}_{0.116}\text{Co}_{0.384}\text{Mn}_{0.5}\text{O}_2$ and (b): $\text{Na}_{1.5}\text{Ni}_{0.117}\text{Co}_{0.366}\text{Al}_{0.017}\text{Mn}_{0.5}\text{O}_2$.

4. Conclusions

$[(1-x-y)\text{NaNi}_{0.7}\text{Co}_{0.3}\text{O}_2]$ doped with Al for cathode materials have been successfully synthesized via a sol-gel method. The results show that all the prepared “ $\text{Na}_{1.5}\text{Ni}_{0.117}\text{Co}_{0.366}\text{Al}_{0.017}\text{Mn}_{0.5}\text{O}_2$ ” type layered structure of the nickel content and Al-doped improve the capacity retention significantly. The percentage of Ni and Co exhibits good efficiency. Although these kinds of systems can help to remove the disadvantage of Co, the efficiency of these kind systems is similar to NaCoO_2 cathode material. Therefore, the fabrication of Sodium-ion batteries using a few more transition elements such as Mn and Al are suggested for any further research.

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Conflicts of Interest

The authors declare no conflict of interest.

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