

Surface Morphology and Crystallinity of Metal Oxides in Nickel-Cobalt Binary System

(Morfologi Permukaan dan Keabluran Oksida Logam dalam Sistem Biner Nikel-Kobalt)

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ABSTRACT

The effect of composition on nickel cobaltites prepared by co-precipitation was investigated. Various compositions were synthesised under different heat treatment conditions. The formation mechanism, lattice parameter and crystallite size were determined by various techniques. X-ray diffraction (XRD) analysis revealed that the optimum condition for the formation of single phase nickel-cobalt oxide (NiCo_2O_4) can be obtained from solution containing Ni: Co ratio of 33.3 mol%: 66.7 mol% with heat treatment at 400 °C. The presence of pure phase NiCo_2O_4 with the particle size of approximately 34.10 nm was also observed in field emission scanning electron microscopy (FESEM). The crystallinity of the synthesized oxides was improved with the increment of Ni content.

Keywords: Composition; co-precipitation; nanosized; nickel-cobalt oxide; phase formation

ABSTRAK

Kesan komposisi ke atas nikel kobaltit yang disediakan melalui ko-pemendakan telah dikaji. Pelbagai komposisi telah disintesis pada keadaan pemanasan yang berlainan. Mekanisme pembentukan, parameter kekisi dan saiz hablur telah ditentukan dengan pelbagai teknik. Berdasarkan analisis pembelauan sinar-X (XRD), keadaan optimum pembentukan fasa tunggal nikel-kobalt oksida (NiCo_2O_4) boleh diperolehi daripada larutan yang mengandungi Ni: Co pada nisbah 33.3 mol%: 66.7 mol% pada suhu pemanasan 400 °C. Kehadiran fasa tulen NiCo_2O_4 dengan saiz zarah lebih kurang 34.10 nm dapat diperhatikan melalui mikroskop elektron imbasan pancaran medan (FESEM). Keabluran oksida yang disintesis meningkat dengan penambahan kandungan Ni.

Kata kunci: Komposisi; ko-pemendakan; nikel-kobalt oksida; pembentukan fasa; saiz nano

INTRODUCTION

The cobaltite system has been the subject of intense research due to its established applications in electrochemistry. It is known that its fundamental and physicochemical properties are dependent on the methods of preparation, composition of oxides and temperature of thermal decomposition. When the size of cobaltite nanoparticles is reduced to the nanometer range, some of their properties can be different compared to samples in micron range. It is believed that by mixing two or more individual oxides together will lead to synergistic effects that may help in the enhancement of the performance of the synthesized materials. This helps to densify the oxides with lower sintering temperature as well as to enhance the grain growth (Brito et al. 2010).

Nickel cobaltite, NiCo_2O_4 is one of the promising metal oxides in the family of cobaltite materials which has a spinel structure AB_2O_4 , with the nickel ions reside at A-sites and cobalt ions at B-sites. The spinel structure has received much attention by numerous researchers as more than 30 ions with radii ranging from 0.5 to 1.0 Å can be incorporated in the spinel-like phases (Rao & Raveau et al. 1998). The metallic ions occupy one-eighth of the tetrahedral interstices and half of the octahedral interstices

in this cubic structure ($a \approx 8 \text{ \AA}$). The nominal composition, $\text{Ni}_x\text{Co}_{1-x}\text{O}_y$ appeared to be one of the promising candidates to be utilised as electrode material in sodium and sodium-ion cells as well as electrocatalyst in advanced alkaline water electrolyzer due to its high electrical conductivity and desirable optical properties in the infrared regions (Owings et al. 2005). However, there appear limited studies on NiCo_2O_4 for energy storage in supercapacitors. It is highlighted that incorporation of nickel into cobalt oxide would further enhance the electrical conductivity of the prepared samples (Tareen et al. 1984; Tharayil et al. 2007).

Researchers have given a considerable attention in synthesizing cobaltite system by exploring the precursors used, preparation methods, processing control and firing temperatures (De Faria et al. 1998; Lapham & Tseung 2004; Roginskaya et al. 1997; Swathi & Buvaneshwari 2008; Tareen et al. 1984). With all these ideas and concepts in mind, researchers have tried to prepare various metal oxides such as nickel ferrite (Duque et al. 2007; Ziemiak et al. 2007), cobalt ferrite (Liu et al. 2005), zinc ferrite (Hakim et al. 2011), zinc cobaltite (Karthikeyan et al. 2009;

Song et al. 2008; Wei et al. 2007), zinc aluminate (Wei & Chen 2006) and so on. There is also a growing interest in synthesizing ternary oxides for diverse applications due to their impressive electrical, magnetic and optical properties like Co-Ni-Fe oxide (Mathe & Sheikh 2010; Kambale et al. 2009), Co-Mg-Al oxide (Ahmad et al. 2011), Mn-Ni-Co oxide (Durán et al. 2005; Peña et al. 2005; Wang et al. 2007) and many more. To date, various transition metals have been used as supercapacitive materials like cobalt oxide (Bahlawane et al. 2007), nickel oxide (Cheng et al. 2006), manganese oxide (Chang et al. 2009; Nayak & Munichandraiah 2009; Yuan et al. 2008), manganese-nickel oxide (Chen & Hu 2003) and others.

A wide option of preparative methods can be employed to obtain the desired, novel products. Some of the methods applied are traditional ceramic preparation or better known as solid-state route and chemical techniques such as sol-gel, electrochemical, solvothermal, hydrothermal, combustion and co-precipitation. Thermal treatment of co-precipitated precursors is proven to be the most promising method in preparing cobaltite spinels (Klissurski & Uzunova 1994). This method offers lower firing temperature, homogeneous product with controlled morphology and high surface area (Bo et al. 2004). In this study, we report on the compositional effect on phase formation and compatibility, surface morphology and particle size of the prepared samples.

MATERIALS AND METHODS

PREPARATION OF THE OXIDES

Oxides with different cation ratios in Ni-Co system (0 mol% Ni: 100 mol% Co, 22.2 mol% Ni: 77.8 mol% Co, 33.3 mol% Ni: 66.7 mol% Co, 55.6 mol% Ni: 44.4 mol% Co, 77.8 mol% Ni: 22.2 mol% Co and 100 mol% Ni: 0 mol% Co) were prepared by co-precipitation method. Nickel and cobalt acetates were mixed with oxalic acid. The oxalic acid was from Alfa Aesar whereas metal acetates used were from Fluka Chemicals and Merck. The precipitates were dried on a hotplate before calcination in a temperature range of 400-700 °C.

CHARACTERIZATION OF THE OXIDES

Phase purity and identification of the samples were examined using Shimadzu XRD-6000 X-ray diffractometer (XRD) with $\text{CuK}\alpha$, $\lambda = 1.5418 \text{ \AA}$ at a scan rate of $2\theta = 2$ degree min^{-1} . The collected data were then refined using ChekCell software for lattice parameter determination. Surface morphology and particle size of the samples were determined from field emission scanning electron microscopy (FESEM) using JEOL JSM 6700F instrument operated at 15 kV while the stoichiometric characteristic was proven from energy dispersive X-ray (EDX) analysis.

RESULTS AND DISCUSSION

The X-ray diffractograms of the prepared NiO, Co_3O_4 and NiCo_2O_4 are shown in Figure 1. Broadening of the diffraction line representing NiCo_2O_4 (inset in Figure 1) is probably due to the formation of small crystallites (Karolus & Łagiewka 2004; Swathi & Buvanewari 2008) and this could have resulted from the decrease of the particle size of this metal oxide if compared to the two monoxides, NiO and Co_3O_4 . On the other hand, a high intensity diffraction pattern displayed by NiO phase revealed highly crystalline nature of the monoxide (Swathi & Buvanewari 2008). The crystallite sizes were calculated using XRD data based on Debye-Scherrer equation as below:

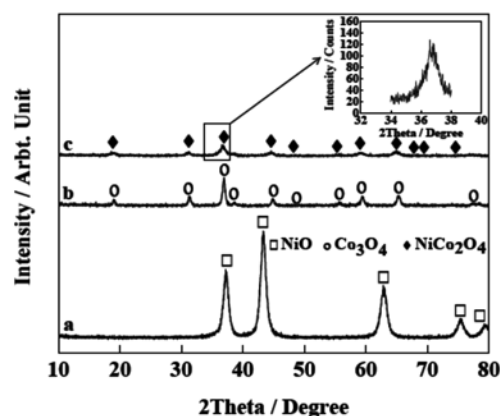


FIGURE 1. XRD patterns of (a) NiO, (b) Co_3O_4 and (c) NiCo_2O_4 (33.3 mol% Ni: 66.7 mol% Co) calcined at 400 °C. Inset is NiCo_2O_4 calcined at 400 °C.

TABLE 1. Crystallite and particle sizes of NiO, Co_3O_4 and NiCo_2O_4 (33.3 mol% Ni: 66.7 mol% Co) calcined at 400 °C

Metal Oxide	Crystallite Size (nm)	Particle Size (nm)
NiO	7.72	42.30
Co_3O_4	14.88	54.43
NiCo_2O_4	10.05	34.10

$$d = \frac{0.89\lambda}{\beta \cos\theta}, \quad (1)$$

where d , λ , θ and β are the crystallite size, X-ray wavelength (1.542 Å), Bragg diffraction angle and full width at the half maximum (FWHM) of the diffraction peak, respectively. The crystallite size and particle size measured from FESEM images are tabulated in Table 1. The crystallite size of NiO was smaller than NiCo₂O₄, but it showed larger particle size in FESEM. This suggested that NiCo₂O₄ was comprised of some or a few small crystallites. On the other hand, NiO has the smallest crystallite size due to its high crystallinity whereas Co₃O₄ gave the largest crystallite size as this compound contains 3 Co ions which has larger radius compared to Ni ion. Figures 2(a) -2(c) show FESEM images of NiO, Co₃O₄ and NiCo₂O₄ at the magnification of $\times 100000$. The particle sizes of NiO and Co₃O₄ were estimated to be 42.30 nm and 54.43 nm, respectively. However, NiCo₂O₄ showed the smallest particle size of 34.10 nm due to the phase purity displayed in XRD as well as the stoichiometric characteristic as observed in EDX with the ratio of the atomic % of Ni: Co is 1.0: 2.1.

The subsolidus solid solution of nickel-cobalt binary system was investigated. The phases present and cell parameters of the prepared samples are listed in Table

2. As shown in Figure 3, NiO becomes the major phase whereas NiCo₂O₄ phase reduced with increasing Ni content. However, the cell parameter increased until a maximum at 66.7 mol% of doped cobalt. The increment is probably associated with the reduction of Ni²⁺ in which smaller Ni²⁺ cations are replaced by larger Co²⁺ cations. The gradual increase is also believed to be resulted from homogeneous precursors via co-precipitation method which lead to complete formation of the spinel phase (Lapham & Tseung 2004). The unit cell of sample with 44.4 mol% of Co decreased with the presence of minute NiO and Co₃O₄ phases. It should be highlighted that there is a close similarity in structure between Co₃O₄ and NiCo₂O₄ (Lapham & Tseung 2004). As Co₃O₄ increased, it become more difficult to distinguish the XRD patterns of these metal oxides.

Figure 4 shows the XRD diffractograms of samples calcined at 700 °C whereas information on lattice parameters and phase purity are tabulated in Table 3. There is no occurrence of spinel NiCo₂O₄ for all compositions. It is understood that NiCo₂O₄ is not stable at temperature above 400 °C which is attributed to the thermal stability limit of this oxide (Peshev et al. 1989). Therefore, samples are then calcined at 700 °C to check on their thermal stability and phase transformation at various cation ratios. High calcination temperature at 700 °C was unfavourable for the formation of NiCo₂O₄ phase as the

TABLE 2. Cation ratio of samples calcined at 400 °C

Cation Ratio Ni: Co (mol%)	Phases Present	Cell Parameter (Å)
0: 100.0	Co ₃ O ₄	8.0835
22.2: 77.8	NiCo ₂ O ₄ and trace NiO and Co ₃ O ₄	NiCo ₂ O ₄ = 8.1116
33.3: 66.7	NiCo ₂ O ₄	NiCo ₂ O ₄ = 8.1295
55.6: 44.4	NiCo ₂ O ₄ and trace NiO and Co ₃ O ₄	NiCo ₂ O ₄ = 8.1142
77.8: 22.2	NiO and trace Co ₃ O ₄ and NiCo ₂ O ₄	
100: 0	NiO	4.1819

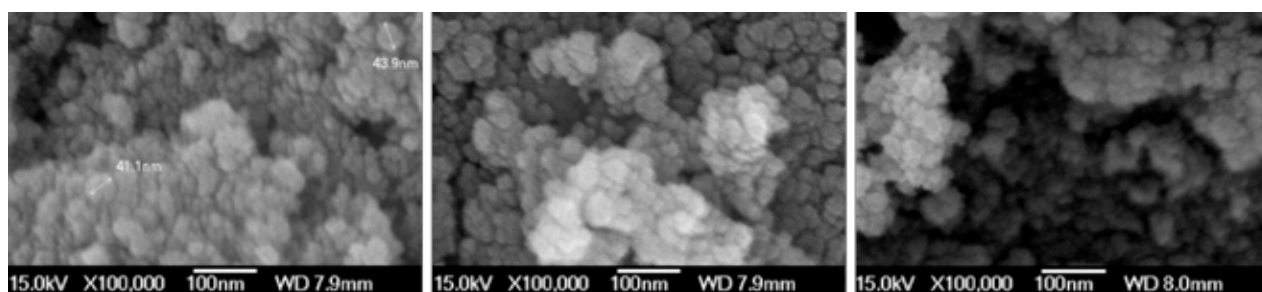


FIGURE 2. FESEM images of (a) NiO, (b) Co₃O₄ and (c) NiCo₂O₄ (33.3 mol% Ni: 66.7 mol% Co) calcined at 400 °C ($\times 100000$ magnification).

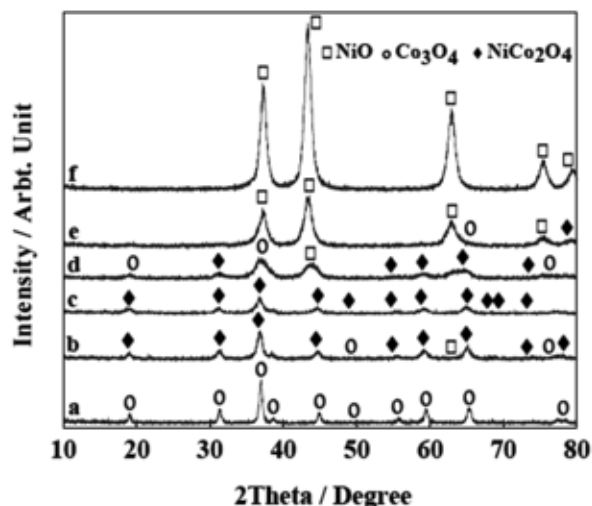


FIGURE 3. XRD patterns of nickel-cobalt oxides calcined at 400 °C: (a) 0 mol% Ni: 100 mol% Co, (b) 22.2 mol% Ni: 77.8 mol% Co, (c) 33.3 mol% Ni: 66.7 mol% Co, (d) 55.6 mol% Ni: 44.4 mol% Co, (e) 77.8 mol% Ni: 22.2 mol% Co and (f) 100 mol% Ni: 0 mol% Co.

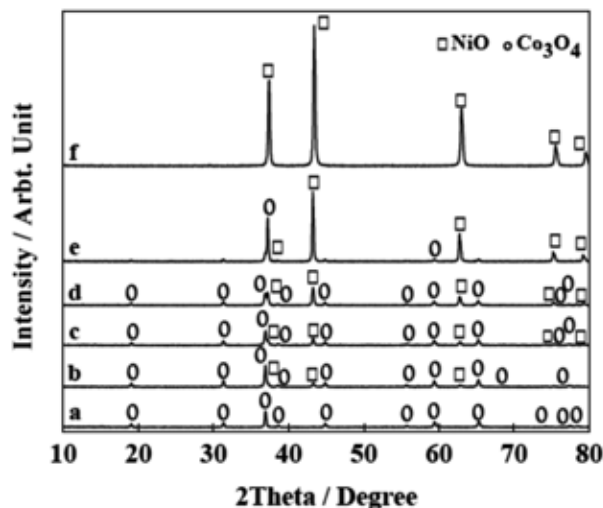


FIGURE 4. XRD patterns of nickel-cobalt oxides calcined at 700 °C: (a) 0 mol% Ni: 100 mol% Co, (b) 22.2 mol% Ni: 77.8 mol% Co, (c) 33.3 mol% Ni: 66.7 mol% Co, (d) 55.6 mol% Ni: 44.4 mol% Co, (e) 77.8 mol% Ni: 22.2 mol% Co and (f) 100 mol% Ni: 0 mol% Co.

TABLE 3. Cation ratio of samples calcined at 700 °C

Cation Ratio Ni: Co (mol%)	Phases Present	Cell Parameter (Å)	Crystallite Size (nm)
0: 100.0	Co ₃ O ₄	8.0853	38.13
22.2: 77.8	NiO and Co ₃ O ₄	Co ₃ O ₄ = 8.0938 NiO = 4.1818	34.07
33.3: 66.7	NiO and Co ₃ O ₄	Co ₃ O ₄ = 8.0963 NiO = 4.1804	32.51
55.6: 44.4	NiO and Co ₃ O ₄	Co ₃ O ₄ = 8.0959	33.25
77.8: 22.2	NiO and Co ₃ O ₄	Co ₃ O ₄ = 8.0895	46.54
100: 0	NiO	4.1751	35.78

samples start to decompose to NiO and Co₃O₄ (Figure 4). However, this has not resulted in any significant changes in the lattice parameter. The crystallinity of the samples improved with higher content of Ni. It was observed that the crystallite size of the prepared metal oxides after heat treatment at 700 °C did not show obvious variation in terms of their crystallite sizes except for 77.8 mol% Ni: 22.2 mol% Co (Ni_{3.5}Co₁O₄) whereby this sample showed better crystallinity with larger crystallite size compared to other cation ratios ranging from 22.2 mol% Ni to 77.8 mol% Ni. Besides, there is a close similarity of crystallite size between 100 mol% Co and 100 mol% Ni at 700 °C. Although the crystallite size of 100 mol% Co was about two times larger than 100 mol% Ni at 400 °C as shown in Table 1, these two samples displayed significant crystallite size enlargement at 700 °C. Therefore, this lead to the interpretation that heat treatment at higher temperature resulted in the formation of larger crystallites for metal oxides.

CONCLUSION

Pure phase spinel structured NiCo₂O₄ was successfully synthesized via oxalate co-precipitation method with cation ratio, 33.3 mol% Ni: 66.7 mol% Co at calcination temperature of 400 °C. This metal oxide possesses particle size of 34.10 nm which was smaller than 100 mol% Co and 100 mol% Ni due to its single phase characteristic. Besides, the stoichiometric feature of this metal oxide was proven in EDX analysis. Increment of cell parameter until a maximum at 66.7 mol% of doped cobalt at 400 °C was due to replacement of Ni²⁺ with larger ionic radius than Co²⁺ as well as the homogeneity ensured by co-precipitation method in producing the spinel oxide. Both NiO and Co₃O₄ phases were observed as secondary phase at other compositions as the formed NiCo₂O₄ was not thermally stable and decomposed at 700 °C. It was found that the crystallinity improved when the content of Ni increased while the crystallite size of the prepared oxides increased with temperature.

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