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Effect of carbon entrapped in Co–Al double oxides on structural restacking and electrochemical performances

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Abstract

Co–Al layered double hydroxides (LDH) were synthesized from nitrates and sodium benzoate by direct coprecipitation, and heated at 600 °C for 3 h in argon gas flow to obtain Co–Al double oxides. The effect of carbon, created during the pyrolysis of benzoate and inserted in resulting double oxides, on structural reconstruction was investigated by X-ray diffraction, scanning electron microscope, Raman spectroscopy, and infrared spectroscopy techniques. It is horizontal arrangement rather than vertical dilayer orientation in the interlayer spacing that was adopted by benzoate.

An abnormal phenomenon was found that when immersed in aqueous 6 M KOH solution in air, the double oxides restacked to Co–Al layered double hydroxides with more regular crystal than before. The reason is believed that carbon was confined in the matrix of resulting double oxides, which prevented further collapse of the layered structure.

Cyclic voltammetries (CV) and constant current charge/discharge measurements reveal that the restacked Co–Al layered double hydroxide has good long-life capacitive performance with a capacitance up to 145 F g^{-1} even at a large current of 2 A g^{-1}. In addition, two clear slopes in chronoampermetric test demonstrated two different diffusion coefficients, explaining the slope of about 118.4 mV in the plot of formal potential E_f versus pOH.
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Keywords: Co–Al layered double hydroxides; Benzoate; Intercalation; Carbon; Supercapacitor; Structural restacking

1. Introduction

Owing to the interlayer space and the anion-exchange ability, layered double hydroxides (LDHs), derived from the brucite $Mg(OH)_2$ and denoted as $[M_{1-x}M_x'(OH)_2](A^{n-})_{x/n}\cdot mH_2O$, are extensively used in catalysis, catalyst support, anion exchange, flame retardants, and slow release of medicine [1–6], where M and M′ stand for divalent and trivalent metallic ions, respectively, and A^{n-} for an interlayer anion. Recently, many new applications have been explored, for example, as catalyst for the creation of carbon nanotubes [7] and template for mesoporous carbon [8]. In electrochemical field, LDHs were tried as electrode active materials for supercapacitor [9–11].

One of the crucial and urgent requirements for supercapacitor in practical applications is high response [12,13], such as in electric vehicles and fuel cell systems. However, to date, little attention [14,15] was put on investigating electrochemical capacitive performances at large current. An obstacle is the deformation and collapse of the structure of electrode material during charge/discharge circling at large current. One of the advantages of using Co–Al LDHs as electrode material is that the structure can be stabilized by partial isomorphous replacement of the cobalt active sites with Al, resulting in an enhancement of Co utilization ratio over Co_3O_4. Wang [11] found that Co–Al LDHs after heating at 160 °C exhibited good supercapacitor behavior due to more exposure of Co active site. Yet, when heating at higher temperature, double oxides obtained from Co–Al LDHs revealed poor electrochemical performances. One of the reasons is the collapse of the layered structure of Co–Al LDHs when calcination temperature is elevated.

Layered double hydroxides is a sort of lamellar structure in which brucite-like host layer charge positively because of partial substitution of divalent metallic ions with trivalent ones. In order to balance excess charges, anions occupy the interlayer spaces with water molecules together. LDHs have an interesting "memory effect" [16–18]. Namely, layered double oxides

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(LDO) can be acquired from layered double hydroxides and LDO can restack to the primary structure. A general view of point is accepted that LDH's structure will collapse if heating is above the temperature at which the formation of spinel phase takes place, and that the resulting double oxides cannot reconstruct to their original architecture again. Therefore, in order to get good structural stability and long-life electrochemical performances, it is necessary to modify or decorate the architecture of Co–Al LDHs.

The physical and chemical properties of layered double hydroxides can be modified and improved by intercalating guest molecules into the gallery spaces. For example, manganese oxide species have been tried to intercalate in Mg–Al layered double hydroxides [19]. Recently, Leroux and coworkers [8] have prepared mesoporous carbon for electric double layer capacitor by pyrolyzing organo-modified layered double hydroxides and dissolving the inorganic moiety, which may provide a new method for preparing carbon-embedded double oxides. Moreover, the carbon confined in LDO may have effects on LDO's reconstruction and electrochemical performances. Taking above facts in mind, in this research, we report the preparation of Co–Al LDHs intercalated by benzoate in the interlayer spaces. Our intention is to obtain carbon-inserted double oxides after the decomposition of Co–Al LDHs in Ar atmosphere due to the pyrolysis of benzoate, and to expect it will maintain the layered architecture. XRD, FTIR, and SEM measurements were employed to valuate the restacking of Co–Al LDHs from carbon-embedded LDO. The electrochemical capacitive performance, long-life stability, and kinetic mechanism of Co–Al LDHs in alkaline solution have also been investigated.

2. Experimental

All chemicals are AR grade and used as received. The synthesis of Co–Al LDH(BA) was carried out by a direct coprecipitation method similar to that described by Prevot [20]. A mixture of $Co(NO_3)_2$ (0.67 M) and $Al(NO_3)_3$ (0.33 M) aqueous solution 20 ml was added dropwisely to 75 ml of a sodium benzoate solution of 0.5 M. Meanwhile, the pH was adjusted at a constant value of 11.0 by adding 1 M NaOH aqueous solution. Then, the mixture was aged at room temperature for 36 h under ambient atmosphere. After filtered and washed with ultrapure water until the pH value of the filtrate was below 8, the residue was air dried at room temperature. The as-prepared Co–Al LDH(BA) was heated at 600 °C for 3 h (heating rate of 10 °C min^{-1}) under continuous argon gas flow in a tubular furnace and cooled down to room temperature, and the product was denoted as Co–Al LDO. After that, Co–Al LDO was immersed in 6 M KOH solution in air for 5 days and filtered. The resulting solid was washed with ultrapure water and anhydrous ethanol for several times, and finally dried in vacuum at 80 °C for 2 h, denoted as Co–Al LDH(OH).

Phase purity of the synthesized materials was studied by X-ray diffraction (Bruker D8 advance, Germany), using Cu Kα radiation (λ = 0.15418 nm). The particle morphology and structural properties of the samples were analyzed by a scanning electron microscopy (FEI Quanta 2000) operated at 20 kV. Fourier transform infrared spectrometer and Raman microscope were used to confirm the intercalation of benzoate in Co–Al LDH and the formation of carbon in Co–Al LDO.

All electrochemical performances were carried out in a conventional three-electrode system equipped with a Pt foil of 1 cm^2 and a saturated calomel electrode (SCE) as counter and reference electrodes, respectively. The working electrode was made from mixing of active material (Co–Al LDH(BA), Co–Al LDO, or Co–Al LDH(OH)), acetylene black, and PTFE with a weight ratio of 80:15:5, coating on a piece of foamed nickel of about 1 cm^2, and pressing to be a thin foil at a pressure of 20 MPa. The electrolyte used was 1 M KOH solution. The cut-off voltage was between −0.2 and 0.6 V versus SCE for cyclic voltammetry (CV) and −0.2–0.55 V versus SCE for chronopentiaometric test. Chronoamperometric measurement was applied at 0 V versus SCE in 1 M KOH solution with a step of 0.6 V. All above tests were carried out on CHI 660B electrochemical workstation system.

3. Results

3.1. Characterizations

Prevot [20] pointed out that Co–Al LDH(BA) could not be acquired from standard anion exchange. So, we synthesized benzoate-intercalated Co–Al LDH by direct coprecipitation and heated it at 600 °C under Ar atmosphere in order to harvest carbon-intercalated LDO. X-ray diffraction pattern of Co–Al LDH(BA) is given in Fig. 1. All reflections in pattern (a) can be indexed to a typical hydrotalcite-like structure (JCPDS: 38-0487). No peaks for impurities are discerned, indicating the purity of Co–Al LDH from direct coprecipitation. The intensities of the peaks for LDHs are not strong enough, implying that the Co–Al LDH is not well crystalline. In pattern (b), all diffraction peaks assigned to layered structure vanish. The weak peaks, occurring at 2θ values of 36.8°, 42.94° and 61.3°, can be indexed to [1 1 1], [2 0 0] and [2 2 0] planes of CoO phase (JCPDS: 43-1004). It consists with the precious report [21] that the CoO phase dispersed on an amorphous $CoAl_2O_4$ phase after decomposition of Co–Al LDH in N_2. With regards to pattern (c),

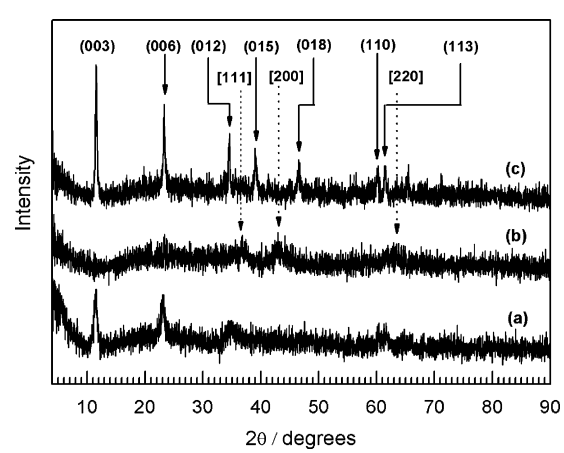

Fig. 1. XRD patterns of Co–Al LDH(BA) (a), Co–Al LDO (b) and Co–Al LDH(OH) (c), in which [ ] and ( ) stand for CoO and Co–Al LDH, respectively.

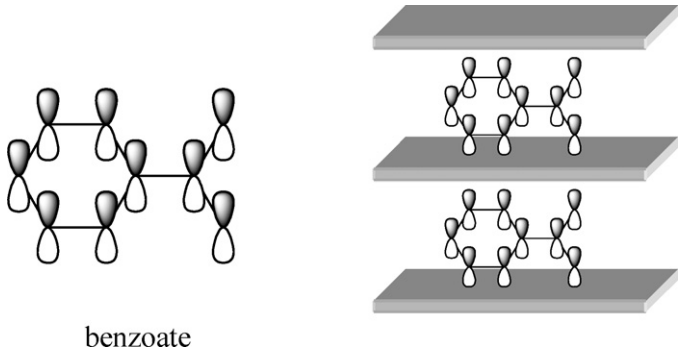

Fig. 2. Schematic representation of Co–Al layered double hydroxide structure consisting of benzoates in the interlayer region.

all peaks assigned to LDHs appear again and the reflections are sharper and more intensive than Co–Al LDH(BA), showing that the restacked Co–Al LDH(OH) has better crystalline structure.

However, the basal spacing value (d_{003} = 1.59 nm) reported in literatures [20,22], corresponding to dilayer benzoate arranged in the LDH interlayer space, was not observed in our experiment. The value of d_{003} for Co–Al LDH(BA) is 0.771 nm which is slightly bigger than that of 0.765 nm in LDHs inserted by carbonate [23]. In previous works, it has been reported that there are two extreme orientations of benzoate [22]: vertical (i.e., perpendicular) and horizontal to the layers. When benzoate adopts vertical orientation, the vertical arrangement is a dilayer whereas the horizontal a monolayer. It was also reported [24] that the angle of the plane of the benzoate molecule to the (0 0 l) planes could change from $35 \pm 10°$ to $0 \pm 10°$. So, we guess that benzoate here maybe adopt a horizontal or declining orientation in our Co–Al LDH(BA). Owing to the conjugation of phenyl with carboxyl groups in benzoate, the electron cloud of the big-π bond is apt to adopt horizontal arrangement in the interlayer spacing of Co–Al LDHs in order to balance well the positive slabs, as shown in Fig. 2.

Lattice parameters [25] c, calculated from $c = 3/2$ ($d_{003} + 2d_{006}$), and a, from $a = 2d_{110}$, and the basal spacing d_{003} are listed in Table 1. The parameter a, the average cation–cation distance within the layers, and c, corresponding to the Coulomb force between the layer and anion in interlayer, are both same as each other, indicating retaining of the layered structure of Co–Al LDHs before and after restacking. Basal spacings of d_{003} are 0.771 and 0.764 nm, respectively. Presumed that the width of the brucite-like layer is 0.48 nm [25], the gallery height is 0.291 nm for Co–Al LDH(BA) and 0.284 nm for Co–Al LDH(OH).

The insertion of benzoate in Co–Al LDHs can be confirmed by infrared spectroscopy (see Fig. 3). For sample (a), the broad peak centered at 3433 cm^{-1} is ascribed to the O-H stretching

Table 1
Lattice parameters (nm) of Co–Al LDHs before and after restacking

Sample	d_{003}	d_{006}	d_{110}	a	c
Co–Al LDH(BA)	0.771	0.385	0.153	0.306	2.312
Co–Al LDH(OH)	0.764	0.381	0.153	0.306	2.289

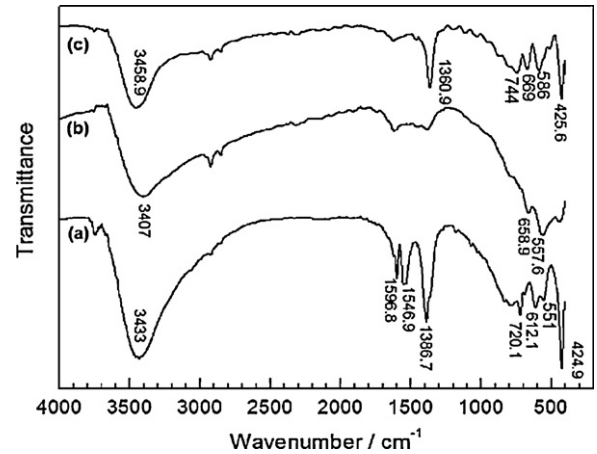

Fig. 3. FTIR spectra of Co–Al LDH(BA) (a), Co–Al LDO (b) and Co–Al LDH(OH) (c).

vibration of water molecules in the interlayer and H-bonded hydroxyl group [26]. The characteristic bands for carboxyl and phenyl groups at 1596, 1546, and 1386 cm^{-1} are clearly observed [20,22], companied with vibration bands for phenyl group at 720 and 833 cm^{-1}. The peaks appeared at 612.1, 551, and 425 cm^{-1} are typical for Co–Al LDHs [11]. The absorption peaks at 1363.5 and 784.9 cm^{-1} are assigned to the vibration of carbonate. Namely, carbonate and benzoate were embedded in the interlayer spaces of Co–Al LDHs together. After sintering at 600 °C in Ar gas, the absorption bands corresponding to carbonate and benzoate both disappear, and the strong broad peak at about 3407 cm^{-1} still appears, indicating that not all the layered structure collapsed. Compared with sample (a and b), the sharp peak at 1360.9 cm^{-1} appears again in sample (c), showing that carbonate was intercalated into the interlayer of Co–Al LDHs after restacking in 6 M KOH solution in air. The FTIR observations are in good accordance with XRD results. So, after a circle of sintering and restacking, Co–Al LDHs inserted with carbonate was obtained once more. During the process, carbon, created at the stage of the pyrolysis of Co–Al LDH(BA), was entrapped in the resultant double oxides, which may prevent complete collapse of layered architecture. Accordingly, the abnormal phenomenon of reconstruction from LDO after calcinations of Co–Al LDH(BA) even at 600 °C for Co–Al LDHs can get explanation.

Fig. 4 provides SEM images of Co–Al LDH(BA), Co–Al LDO, and Co–Al LDH(OH). As can be seen, Co–Al LDH(BA) has layered structure despite that it is not very uniform. Co–Al LDO reveals no clear laminative fabrication, but it still maintains somewhat layered profile if observed carefully. With regards to Co–Al LDH(OH), the sample consists of well-crystalline thin platelets with a thickness of about several nanometers. The observation from SEM images of three samples is also well consistent to XRD results.

3.2. Electrochemical properties

Constant current charge/discharge curves at different current density for three samples in 1 M KOH solution were presented in Fig. 5. The cut-off voltage is between −0.2 and 0.55 V ver-

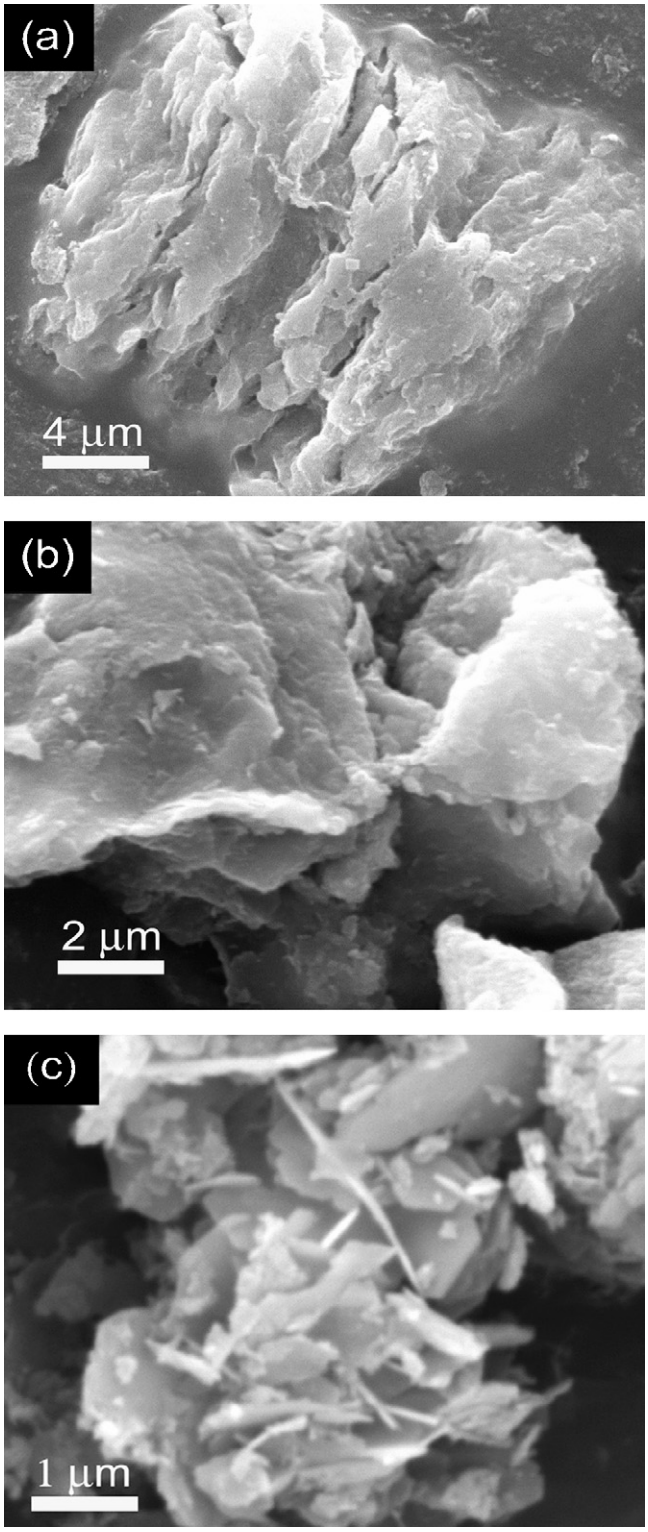

Fig. 4. SEM images of Co–Al LDH(BA) (a), Co–Al LDO (b) and Co–Al LDH(OH) (c).

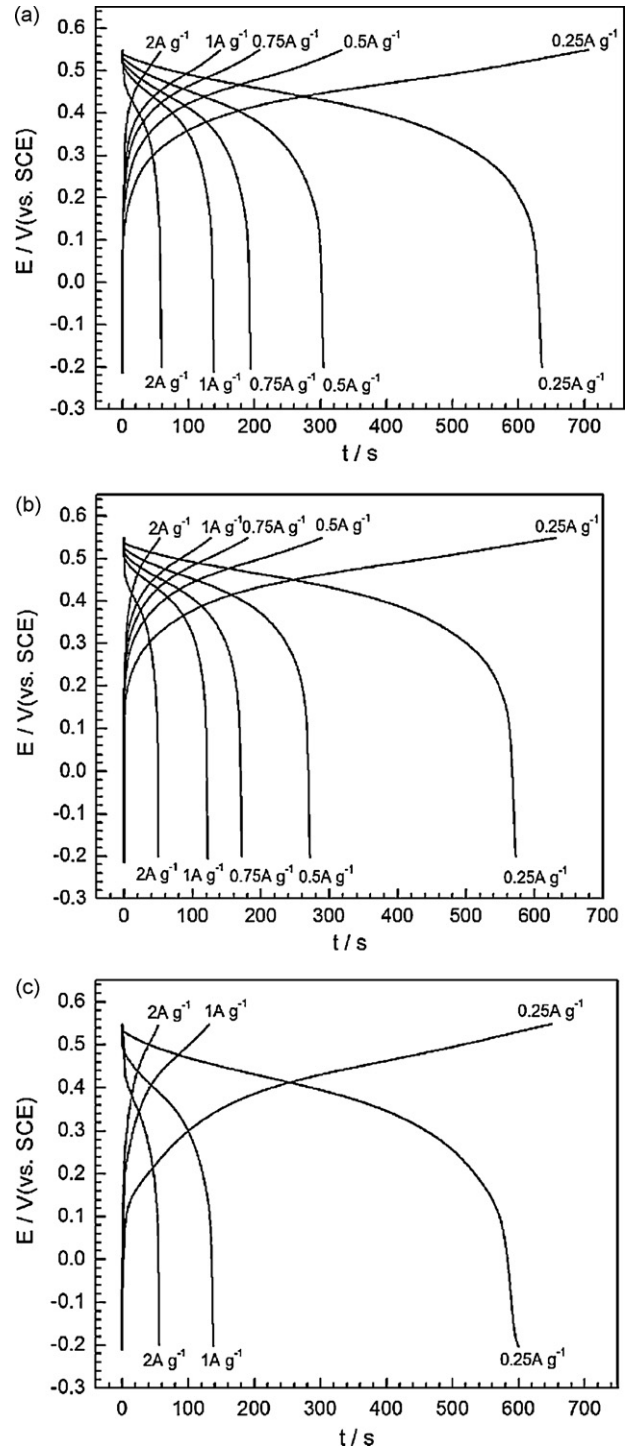

Fig. 5. Charge/discharge curves of Co–Al LDH(BA) (a), Co–Al LDO (b) and Co–Al LDH(OH) (c) at various current densities.

sus SCE. As shown in Fig. 5, the three samples have some common: charge/discharge time decreases with specific current. Corresponding capacitances calculated from Eq. (1) are listed in Table 2, where I stands for charge or discharge current, Δt for charge or discharge time, m for the mass of the electrode, and ΔV stands for the potential range during charging or discharging.

$$C_s = \frac{I\,\Delta t}{m\,\Delta V} \quad (1)$$

When specific current is $0.25\,\mathrm{A\,g^{-1}}$, the specific capacitances (C_s) of the three samples are between 190.9 and $212.1\,\mathrm{F\,g^{-1}}$. When current increases to $2\,\mathrm{A\,g^{-1}}$, C_s is 132.3–$157.9\,\mathrm{F\,g^{-1}}$. Namely, no great difference in C_s comes forth as discharge cur-

Table 2
Charge/discharge capacitances of the three samples at various current densities

Current ($A\,g^{-1}$)	Co–Al LDH(BA)		Co–Al LDO		Co–Al LDH(OH)	
	Charge ($F\,g^{-1}$)	Discharge ($F\,g^{-1}$)	Charge ($F\,g^{-1}$)	Discharge ($F\,g^{-1}$)	Charge ($F\,g^{-1}$)	Discharge ($F\,g^{-1}$)
0.25	235.5	212.1	216.7	199.9	210.6	190.9
0.5	221.5	203.1	–	–	192.6	181.3
0.75	207.1	194.2	–	–	180.5	172.1
1	197.2	185.6	131.5	138.2	169.1	162.8
2	152.6	157.9	145.1	149.3	138.7	132.3

rent increases from 0.25 $A\,g^{-1}$ to a large current of 2 $A\,g^{-1}$. This indicates that three samples have the ability of charge/discharge at large load and are promising candidates for electrode materials of supercapacitor. It is also can be confirmed by cyclic voltammetries at various scan rates (see Fig. 6) that the current is directly proportional to scan rate.

In order to have a good understanding of long-life capacitive properties of the three samples, we carried out charge/discharge cyclic measurements at a large current of 2 $A\,g^{-1}$ between −0.2 and 0.55 V versus SCE in 1 M KOH solution. Fig. 7 gives the relation of C_s to cyclic numbers. For Co–Al LDH(BA), starting specific capacitance is high up to 153.6 $F\,g^{-1}$ and can be maintained for about 100 cycles. Then C_s decreases with cycle to a value of 132.3 $F\,g^{-1}$ at the 1000th cycle with a capacitance retention of 86.1%. Co–Al LDO has a specific capacitance of 132.3 $F\,g^{-1}$ for the first cycle and declines acutely during the first 100 cycles, and then decreases slowly. After 781 cycles, its capacitance is 123.7 $F\,g^{-1}$ with retention of 93.5%. As for Co–Al LDH(OH), its performance is very different from the first two samples. From 1 to 250 cycles, C_s keeps 145 $F\,g^{-1}$. Then C_s increases, maintains 149.3 $F\,g^{-1}$ until the 900th cycle, and decreases to 145 $F\,g^{-1}$ finally. So, its retention of capacitance is 100%, showing good long-life stability at heavy current density.

4. Discussions

It is interesting to notice that Co–Al LDH(OH) restacked from Co–Al LDO holds better capacitive properties in 1 M KOH aqueous solution than Co–Al LDH(BA). From XRD patterns and SEM images, it can be found that Co–Al LDH(OH) not only can be restacked from LDO which was obtained from Co–Al LDH(BA) after sintering at 600 °C, but also has more regular layered structure than Co–Al LDH(BA). The fact conflicts with common viewpoint that LDO obtained from calcinations of LDHs above 500 °C cannot reconstruct to LDH again because of the collapse of the layered architecture. It has been reported [27] that the lamellar structure of Co–Al LDH intercalated with acetate collapses and the diffraction lines of the Co_3O_4 and $CoAl_2O_4$ spinel phases appear before 250 °C. When Co–Al LDH was heated at 250–300 °C in air, complete dehydroxylation leaded to the collapse of the hydrotalcite phase. When in inert gas, the temperature is 350–400 °C [21]. A reasonable explanation here may be that carbon was created during carbonization of benzoate at 600 °C under Ar atmosphere and confined in the resulting LDO, which prevented further collapse of layered structure. So, Co–Al LDH is formed once more when carbon-containing LDO is dipped in 6 M KOH solution. The hypothesis can be confirmed by Raman spectra in Fig. 8. When Co–Al LDO was dissolved in 0.5 M H_2SO_4 solution, filtered and washed by ultrapure water, a black powder was obtained. Two main peaks at 1344 and 1563.9 cm^{-1} appear in its Raman spectrum as expected for carbon material [8], corresponding to the D and G bands of carbon, respectively. This proves our primary intention that benzoate was inserted in the interlayer spacing of Co–Al LDH by direct coprecipitation, and that carbon was formed when Co–Al LDH was heated at 600 °C under Ar gas flow.

Another interesting phenomenon is the redox potentials for Co(+2)/Co(+3) reaction for three samples in 1 M KOH solu-

Table 3
Relation of formal potentials of the three samples to concentration of OH^- anion

Sample	C_{OH^-} (M)	$E_{a,p}$ (V)	$E_{c,p}$ (V)	E_f (V)	Slope (mV)	Relative coefficient
Co–Al LDH(BA)	0.5	0.4568	0.3660	0.4114	120.63	0.93888
	0.67	0.4492	0.3668	0.4080		
	0.8	0.4266	0.3598	0.3932		
	1.0	0.4072	0.3440	0.3756		
Co–Al LDO	0.5	0.4652	0.3662	0.4157	117.58	0.99195
	0.67	0.4428	0.3632	0.4030		
	0.8	0.4208	0.3582	0.3895		
	1.0	0.4172	0.3458	0.3815		
Co–Al LDH(OH)	0.5	0.4038	0.3451	0.3744	121.62	0.99424
	0.67	0.3860	0.3246	0.3553		
	0.8	0.3728	0.3236	0.3482		
	1.0	0.3662	0.3088	0.3375		

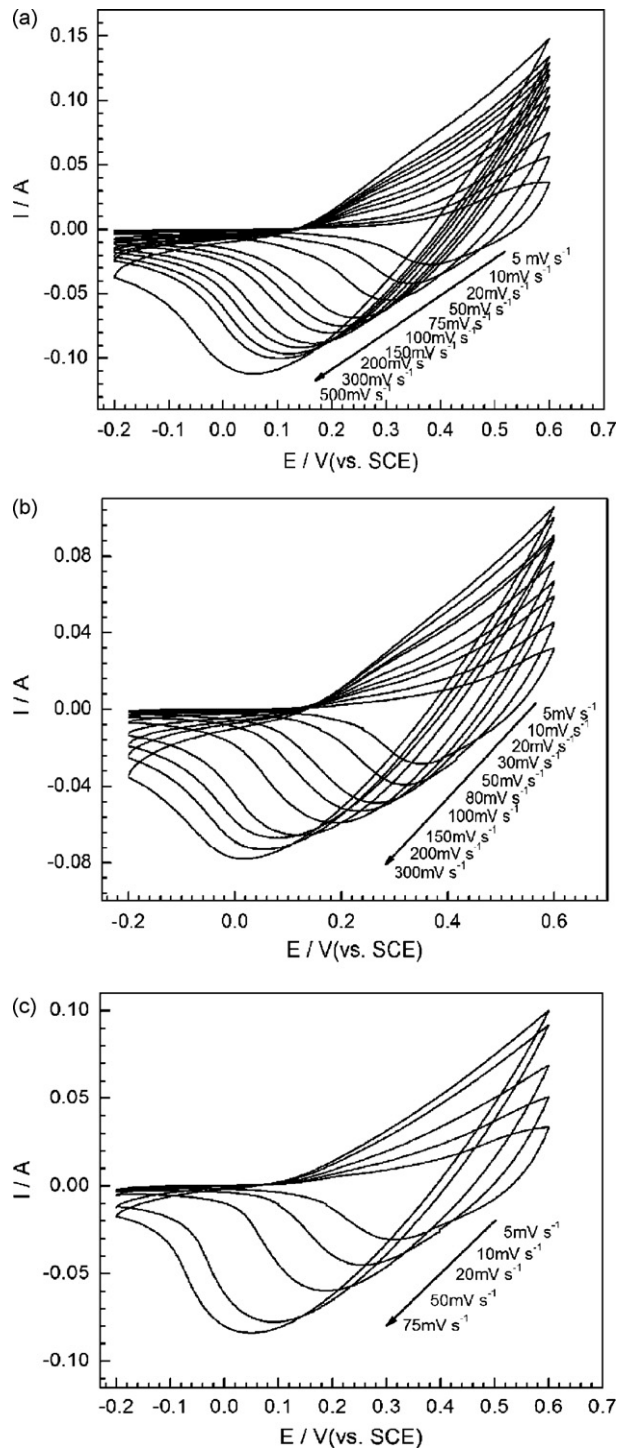

Fig. 6. Cyclic voltammograms of Co–Al LDH(BA) (a), Co–Al LDO (b) and Co–Al LDH(OH) (c) at various scan rates.

tion in CV curves. For simplicity, only CV curves at a scan rate of 0.5 mV s^{-1} are displayed in Fig. 9. It has been reported [7] that Co–Al LDH and Co–Al LDO acquired from Co–Al LDH after annealing at various temperature have different anodic and cathodic peak potentials in CV curves. However, in our experiments, to our surprise is that the potentials corresponding to Co(+2)/Co(+3) conversion appear at almost the same position, meaning that Co–Al LDH(BA), Co–Al LDO, and

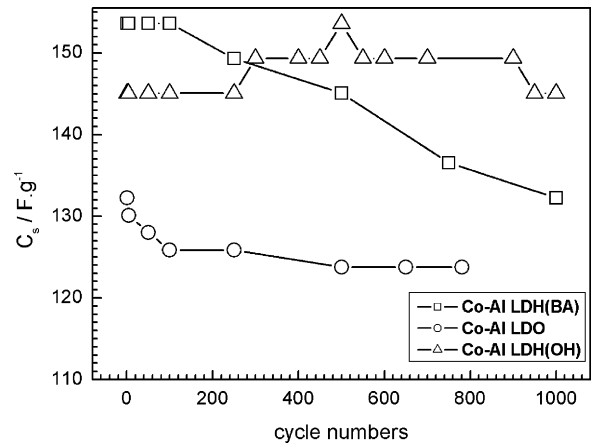

Fig. 7. The relation of weight-specific capacitance of the three samples to charge/discharge cyclic number.

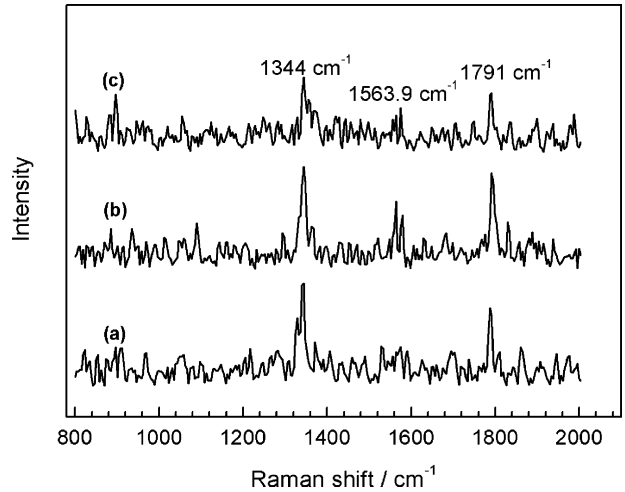

Fig. 8. Raman spectra of acetylene black (a), graphite (b), and carbon from dissolving Co–Al LDO (c).

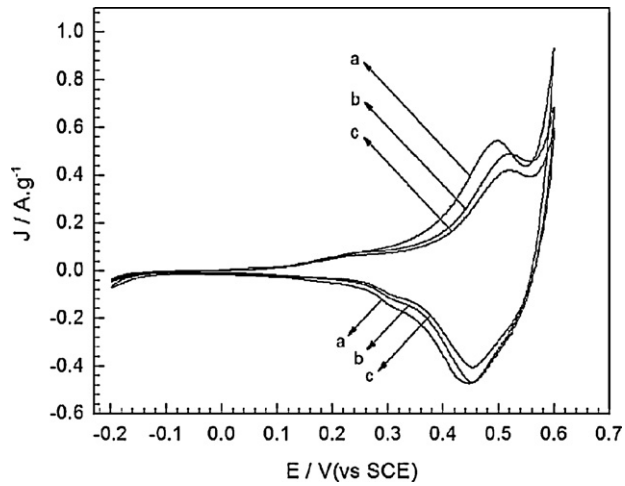

Fig. 9. Cyclic voltammograms of Co–Al LDH(BA) (a), Co–Al LDO (b) and Co–Al LDH(OH) (c) at a scan rate of 0.5 mV s^{-1}.

Co–Al LDH(OH) share the same mechanism of redox reaction. The formal potential, E_f, calculated [28] approximately from $1/2(E_{a,p} + E_{c,p})$, at various concentration of KOH, are listed in Table 3, where $E_{a,p}$ and $E_{c,p}$ are the anodic and cathodic peak potentials in CV curve. According to Eq. (2), investigation on the relation of E_f with pOH helps to understand the role of OH^- ion in the reaction.

$$E_f = E_0' + \frac{RT}{nF} \ln c_{OH^-}^m \quad (2)$$

The dependence of formal potentials for the three samples on the values of pOH is showed in Fig. 10 in order to have a clear observation. The excellent linear correlation of E_f to pOH indicates that OH^- anion takes part in the reaction of conversion between Co(+2) to Co(+3). However, there are some differences between Scavetta et al.'s observation [29] and ours. In their report, the slope was 59.2 mV, indicating single OH^- ion participating in the reaction ($m = 1$), whereas it is very near to 118.4 mV in our tests, which means that two OH^- ions ($m = 2$) participate the reaction of Co(+2)/Co(+3) because only one electron is transferred in the process. In the beginning, we have ever doubted that K^+ ion takes part in the process with OH^- ion together. But there is not any redox wave at all when the same electrode was soaped into 0.375 M and 0.5 M K_2SO_4 solutions. So, the probability from K^+ ion can be excluded.

We also carried out a chronoamperometric measurement and found two clear slopes in the plot of $I(t)$–$t^{1/2}$, indicating two apparent diffusion coefficients of OH^- ion (see Fig. 11). Considering the low concentration of OH^- ion used in their report and high concentration in our experiments, we propose an explanation as follows: In the whole process of transformation from Co(+2) to Co(+3), OH^- ion must come through two steps. One is the diffusion of OH^- ion from bulk solution to the surface of LDH; the other is the diffusion of OH^- ion from the surface to the bulk of solid LDH, companied with the transfer of one electron. It can be written as:

$$OH_{sol}^- \xrightarrow{\text{diffusion}} OH_{sur}^- \quad (3)$$

$$\text{HT–Co(+2)} + OH_{sur}^- \xrightarrow{\text{diffusion}} \text{HT(OH)–Co(+3)} + e^- \quad (4)$$

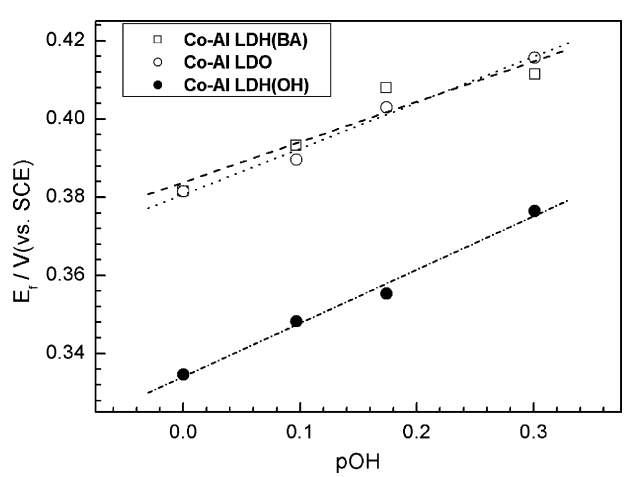

Fig. 10. Dependence of formal potentials for the three samples on pOH.

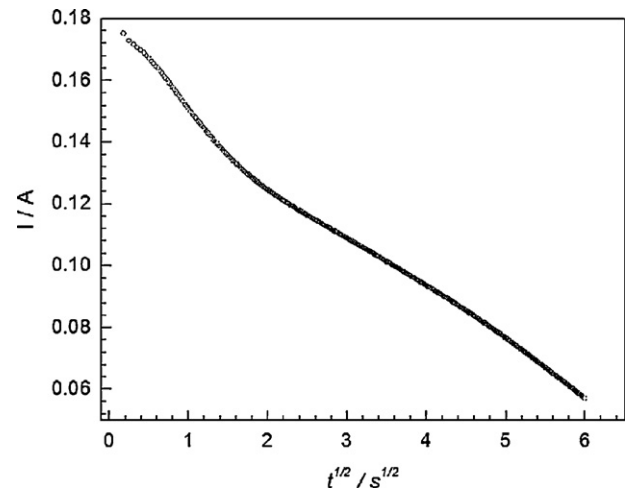

Fig. 11. Plot of $I(t)$ vs. $t^{1/2}$ for Co–Al LDH(OH), operating in 1 M KOH solution, $E_{in} = 0$ V, $E_{step} = 0.6$ V.

where HT means hydrotalcite. OH_{sol}^- and OH_{sur}^- stand for OH^- ions in the bulk solution and on the surface of LDH, respectively. When the solution is weakly basic, there is no great difference in the concentration of OH^- ion between on the surface of LDH slat and in the bulk solution. The OH^- anions needed for the second step can be easily compensated by *in situ* ionization of water molecules without necessary diffusion of OH^- ions from distant bulk solution. Therefore, the kinetics of OH^- ion is one order. When the concentration of OH^- ion in the solution is high, OH^- ion, created by the ionization of water molecules in the interlayer spacing of LDH, is relatively low. Hydroxyl anions must diffuse from the bulk solution to the surface of LDH slat to supply enough OH^- ions for further diffusion into the bulk of solid LDH. So OH^- ion is kinetically two orders. This explanation can also be applied to the observation for Ni–Al LDH in precious work [30], where the OH^- ion diffusion in the interlayer is the rate-determining step of the reaction.

5. Conclusions

Co–Al layered double hydroxides consisting of benzoate were prepared by a direct coprecipitation. XRD, FTIR, SEM, CV, and constant current charge/discharge measurements were employed to valuate the effect of carbon inserted in LDO on the structure and electrochemical properties. Following conclusions are drawn out:

(a) As-prepared Co–Al LDH(BA) is a lamellar structure, in which benzoate adopts horizontal orientation in the interlayer spacing.
(b) Co–Al LDO from calcinations of Co–Al LDH(BA) holds good "memory effect" even at 600 °C under Ar atmosphere. The restacked Co–Al layered double hydroxides have more regular crystal than before. The explanation for this abnormal phenomenon is possibly that carbon, inserted in Co–Al LDO during the calcination of Co–Al LDH(BA), can prevent LDO from further structural collapsing.

(c) The reconstructed Co–Al layered double hydroxides reveal good long-life cyclicity and high capacitance of 145 F g^{-1} even at a large current of 2 A g^{-1}, which is very possibly related to the structure-supporting effect of intercalated carbon.

(d) Co–Al LDH(BA), Co–Al LDO, and Co–Al LDH(OH) share the same reaction mechanism of two-step diffusions for OH^- anion from the bulk of the electrolyte to bulk slabs of LDHs.

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