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COMMUNICATION

Influence of Temperature and Electrolyte Concentration on the Structure and Catalytic Oxygen Evolution Activity of NiFe LDH

Corina Andronescu[a,b]*, Sabine Seisel[a], Patrick Wilde[a], Stefan Barwe[a], Justus Masa[a], Yen-Ting Chen[a], Edgar Ventosa[c], Wolfgang Schuhmann[a]*

Abstract: NiFe layered double hydroxide is inarguably the most active contemporary catalyst for the oxygen evolution reaction under alkaline conditions. However, the ability to sustain unattenuated performance under challenging industrial conditions entailing high corrosivity of the electrolyte (~30 wt.% KOH), high temperature (> 80 °C) and high current densities (> 500 mA cm^{-2}) is the ultimate criterion for practical viability. Here, we evaluate the chemical and structural stability of NiFe LDH at conditions akin to practical electrolysis, in 30 % KOH at 80 °C, however, without electrochemical polarization, and the resulting impact on the OER performance of the catalyst. Post-analysis of the catalyst by means of XRD, TEM, FT-IR and Raman spectroscopy after its immersion into 30 % M KOH at 80 °C for 60 h revealed a transformation of the structure from NiFe LDH to a mixture of crystalline β-$Ni(OH)_2$ and discrete predominantly amorphous FeOOH containing minor non-homogeneously distributed crystalline domains. These structural and compositional changes were leading to a drastic loss of the OER activity. We therefore recommend studying catalyst stability at industrially relevant conditions.

In recent years, improvement of the activity of catalysts for the oxygen evolution reaction (OER) or developing novel OER catalyst materials especially in alkaline solution were the main research goals in electrocatalysis. The quest of the Grail was to find catalysts with decreased overpotential for the OER usually comparing the catalytic activity at a current density of 10 mA cm^{-2} normalized to the geometric footprint of the electrode without considering the true electrochemically active surface area of the catalyst-modified electrode. Long-term stability measurements typically performed at the same current density and for a rather short duration were already facing difficulties concerning catalyst particle adhesion in the often drop-coated catalyst film. Of all the reported abundant non-noble metal-based OER catalysts, NiFe LDH stands out among the most active candidates.[1,2] NiFe LDH derives its exceptional activity from the unique layered structure. A previous study by Hunter et al. on the influence of structural changes of NiFe LDH catalysts containing different anions on their OER activity in 1 M KOH at room temperature revealed replacement of the different anions by CO_3^{2-} and a concomitant decrease of catalytic activity.[3] The influence of the degree of crystallinity or sheet size[4] of LDH materials on the catalytic OER performance was also addressed.[2] However, no clear correlation between activity, crystallinity or sheet size was established since these parameters usually change concurrently. Despite huge efforts to develop more active catalysts by combining NiFe LDH with different carbon nanomaterials[5] or depositing NiFe LDH on 3D supports to benefit from an increased surface area,[6] little focus was paid on the investigation of the operational stability of these materials. However, considering the development of non-noble metal and abundant highly active OER catalysts as an important contribution to solve limitations in alkaline electrolyzers for water splitting, the knowledge about their intrinsic chemical stability under close to industrial operating conditions is of utmost importance. Aging studies performed on NiFe LDH at moderate temperature in 1 M or 10 M KOH indicated no change in the catalytic activity.[7] The transformation of $Ni(OH)_2$ in KOH, and the influence of the incorporation of Fe impurities on the electrocatalytic OER performance of Ni-based catalyst have been previously reported,[7,8] however, no similar studies exist for NiFe LDH. Recently, we introduced polybenzoxazines as precursor of a carbon matrix, which was successfully used for the stable immobilization of catalyst powders on electrodes.[9] Moreover, due to the high stability and the favourable conductivity, this approach did not only allow to challenge the immobilized catalyst at high current densities and at elevated temperature but also to perform post-mortem analysis of the catalyst layer. With respect to NiFe LDH our results demonstrated that after 100 h of galvanostatic operation at a current density of 200 mA/cm^2 in 5 M KOH at 60 °C, the NiFe LDH was structurally transformed into a mixture of NiO and $NiFe_2O_4$.[10] Evidently, these conditions are still less demanding than the typical conditions of a technical electrolyzer, in which 7.5 M KOH, 80 °C and up to 500 mA/cm^2 current density are applied. Development of both active and stable catalysts for OER is essential for advances in water electrolysis, where high KOH concentrations (~ 30 wt.%), high temperatures (~ 80 °C) and high current densities (~ 500 mA cm^{-2}) are simultaneously applied.[11] However, due to the vigorous gas evolution at both anode and cathode, the corrosive properties of the electrolyte solution and the higher temperature, such an experiment cannot be reliably performed in a three-electrode setup. Preserving a reliable and known reference potential, a secure immobilisation of the catalyst material on the electrode surface as well as conserving the potentiostat feedback loop to function properly considering the high gas bubble load in the solution adding up to a very high uncompensated

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resistance in the electrochemical cell still allowing post electrolysis catalyst characterisation was not performed.

Hence, in order to qualify a novel OER catalyst to be potentially applicable for alkaline water splitting, not only an initially determined low overpotential has to be demonstrated but also the preservation of the structural and compositional features of a catalyst at the aforementioned close to industrial conditions. Considering this, an initially exceptionally active catalyst may turn out to decay fast while a lower initial activity may pay back with enhanced stability under the harsh conditions.

Since it is – as discussed above - experimentally challenging to perform laboratory experiments at these conditions, we propose a straightforward preliminary stability test for potentially interesting OER catalyst by just exposing the catalyst powders to the high-concentrated electrolyte solution at high temperature. In the present work, we investigated the chemical and structural stability of NiFe LDH in KOH of different concentrations and at different electrolyte temperatures over a period of 60 h without applying the high potential necessary to invoke the OER. We consider our study as an example for evaluating the stability of catalyst materials which should be performed as a standard test before claiming high stability and suggesting potential future technical applications. Specifically, the non-electrochemical change in the structure as well as electrochemical activity of NiFe LDH upon exposure to high concentrations of KOH and high temperature is considered to be an additional piece in the puzzle which needs to be solved to completely understand the NiFe LDH system.

We immersed NiFe LDH for 60 h in 7.5 M KOH (~ 30 wt.%) at 80 °C in order to simulate real operating conditions, and as reference, comparative studies were carried out in 1.0 M KOH at 25 °C, which is usually used in laboratory experiments. Powder X-ray diffraction patterns of pristine NiFe LDH, as well as of the samples treated in 1.0 M KOH at ambient temperature and in 7.5 M KOH at 80 °C are shown in Figure 1. The reflection at 2θ = 9.3° is due to the (003) plane and corresponds to the basal spacing. The broad reflection at 2θ = 33.6° originates from the (100) plane while the one at 2θ = 59.9° is attributed to diffractions at the (110) plane. The XRD pattern of pristine NiFe LDH agrees well with reported patterns for layered double hydroxides.[12] However, it has to be noted that the reflections are broad and asymmetric, indicating small domain sizes and a low stacking order.[13] This is further supported by the absence of reflections caused by the (006) plane.[14]

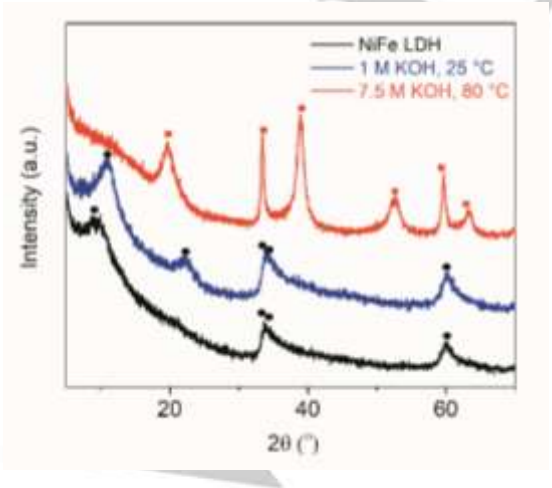

Figure 1. X-ray diffractograms registered for NiFe LDH (black), and NiFe LDH after immersion for 60 h in 1.0 M KOH at 25 °C (blue) and 7.5 M KOH at 80 °C (red) (●- NiFe CO_3^{2-}, ■ β-Ni(OH)$_2$).

After treatment of the sample in 1 M KOH for 60 h at ambient temperature, slight changes can be observed in the X-ray diffractogram. The 003 reflection is shifted to higher values (2θ = 11.2°) due to a decrease in the basal spacing caused by restructuring of the material or replacement of the NO_3^- anions by CO_3^{2-} or OH^- ions.[3,4] A new reflection emerged at 2θ = 22.4°, which is due to the (006) plane of the layered double hydroxide. The changes indicate an increase in the stacking order. Major differences can be observed in the diffraction pattern of NiFe LDH treated in 7.5 M KOH for 60 h at 80 °C. The reflections at 2θ = 11.2° and 22.4° disappeared and new peaks emerged. Compared to the diffraction pattern of the untreated sample, all peaks showed increased intensity. All observed reflections agree with reported values for β-Ni(OH)$_2$ isomorphous with brucite[15] indicating that this is the only crystalline phase clearly detectable by X-ray diffraction. The specific broadening of all non (hk0) reflections indicate interstratification of the β-Ni(OH)$_2$ with α motifs originating from the initial LDH structure or intermediary formed α-Ni(OH)$_2$.[16] No evident structural transformations were observed by XRD when NiFe LDH was immersed in 1.0 M KOH at 80 °C or in 7.5 M KOH at 25 °C (Figure S1), underlining the importance of the conditions that the catalyst is exposed to.

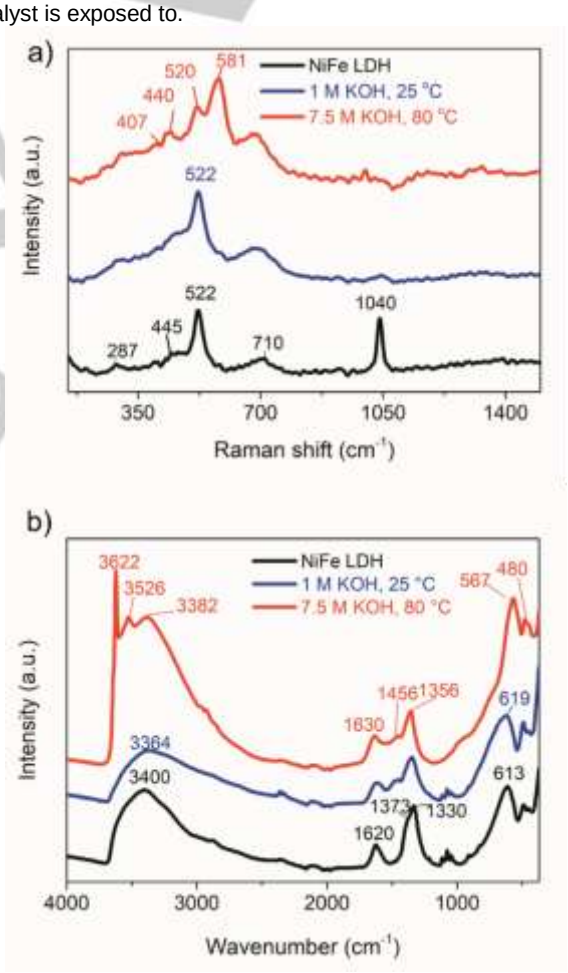

Figure 2. a) Raman and b) FTIR-ATR spectra registered for NiFe LDH (black), and NiFe LDH after immersion for 60 h in 1.0 M KOH at 25 °C (blue) and 7.5 M KOH at 80 °C (red).

The Raman spectrum of the pristine NiFe LDH shows the characteristic peaks reported for brucite like structures at 287, 445 and 725 cm^{-1} (Figure 2a).[17] The intense band at 522 cm^{-1} is also characteristic for brucite type structures and was reported for

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samples which exhibit a low degree of crystallinity.[18] The sharp peak at 1040 cm^{-1} corresponds to the ν_1 nitrate vibration mode. After immersing NiFe LDH for 60 h in 1.0 M KOH at 25 °C, the peak at 1040 cm^{-1} significantly decreases indicating that nitrate ions were replaced by OH^- ions. Besides that, no other modification was observed in the Raman spectrum suggesting that the general structure of LDH is maintained.After immersing NiFe LDH in 7.5 M KOH at 80 °C, significant changes are observed. The peaks at 520 and 440 cm^{-1} are still present. They are attributed to lattice vibrations of β-$Ni(OH)_2$, which is isostructural with the brucite structure of $Mg(OH)_2$ and thus with LDH materials.[19] Additionally, a new band can be observed at 581 cm^{-1}, the intensity of which suggests that it originates from a major compound present in the sample. As reported in literature,[19] Fe can be incorporated into the lattice of β-$Ni(OH)_2$ leading to a disorder which is associated with a blue shift of the peak at 520 cm^{-1}, suggesting that the band at 581 cm^{-1} can be assigned to β-$Ni(OH)_2$ incorporated with Fe.

FTIR-ATR spectra of pristine NiFe LDH and the samples treated in 1.0 M KOH at ambient temperature as well as in 7.5 M KOH at 80 °C are presented in Figure 2b and show the same trend as the X-ray diffractograms and the Raman spectra. The spectrum of the untreated NiFe LDH is in good agreement with reported literature data on LDHs.[20] A broad absorption band of the OH-stretching vibration of layer OH and intercalated water is observed with a maximum at 3400 cm^{-1}, whereas the OH-bending modes of intercalated water can be found at 1620 cm^{-1}. The ν_3 vibration modes of nitrate appear at 1330 cm^{-1}, and below 1000 cm^{-1} the lattice vibrations exhibit maxima at 610 cm^{-1} (M-OH) and 480 cm^{-1} (Ni-O).[20] No major changes occur in the ATR-FTIR spectrum of NiFe LDH after treatment in 1.0 M KOH at ambient temperature. However, similarly as previously noticed in the Raman spectra, the absorption band due to intercalated nitrate ions was not any longer observed. Even though replacement of nitrate by carbonate seems feasible, especially since the treatment was performed under ambient atmosphere, the strong Raman-active ν_1 symmetric stretching vibration of carbonate at 1050 cm^{-1} is not observed. However, at the strong alkaline treatment conditions, a replacement of nitrate by hydroxide is possible as reported by Hall et al.[21] The frequencies of the bending mode of this anion should lie between the one of intercalated water at 1630 cm^{-1} and the lattice vibration of M-OH at 610 cm^{-1}. In fact, these bands were observed between 1380 cm^{-1} and 1480 cm^{-1} in previous studies.[19] The slight changes in shape, blue shift of the maximum to 1356 cm^{-1}, as well as the appearance of a small absorption band at 1456 cm^{-1} support this interpretation. Moreover, in the OH stretching regime a strong peak at 3622 cm^{-1} appears together with a red-shift of the M-OH bending mode to 567 cm^{-1} which indicates the presence of non-hydrogen bonded OH-groups typical for β-$Ni(OH)_2$.[22] In addition, the broad absorption feature around 3400 cm^{-1} is separating into two distinct features, an absorption band at 3526 cm^{-1} assigned to the stretching vibration of hydrogen-bonded OH^- groups and a broader absorption around 3382 cm^{-1} due to the ν_1 vibration of intercalated and adsorbed water.[23] The observed absorption features in the OH-stretching regime indicate interstratification of the β-$Ni(OH)_2$ with α motifs[22] in agreement with the findings of the XRD study.

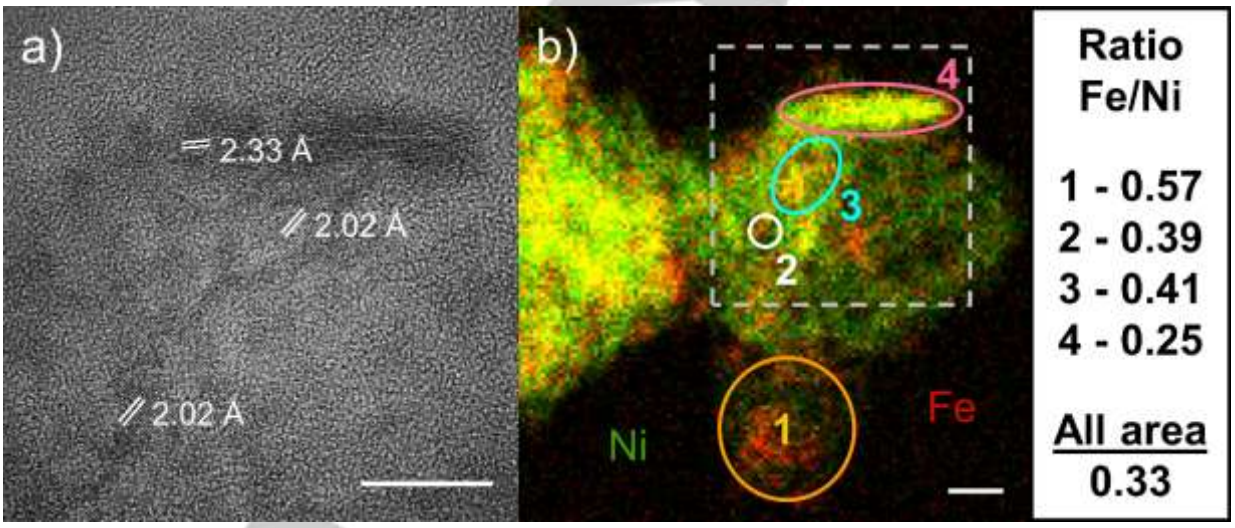

Figure 3. a) HRTEM of NiFe LDH after 60 h immersion in 7.5 M KOH at 80 °C. b) STEM-EDS of the identical location. Signals of Fe (red) and Ni (green) are superimposed. HRTEM outline is indicated by dashed box. The Fe/Ni ratio quantified in the marked areas of Fig. 3b as well as for the entire area is presented in the right part of the figure. Scale bar equals 10 nm in both cases.

No distinct features for an iron-phase could be observed in either Raman, FT-IR spectra or XRD. According to Pourbaix diagrams, Fe should be present in the 3+ oxidation state as a hydroxide in highly alkaline environment.[24] Indeed, EDS-analysis clearly indicates that the Ni/Fe ratio does not change upon alkaline treatment of NiFe LDH (Table S1). This finding implies that iron is not effectively removed from the sample but has to remain present either in form of an incorporation into the lattice of $Ni(OH)_2$ or possibly also in form of amorphous Fe(III) species. HRTEM images (Figure 3a and Figure S2 a and b) show the presence of β-$Ni(OH)_2$ with lattice spacings of 2.33 Å, 2.34 Å and 4.53 Å corresponding to the (101) and (001) planes, respectively.[15] Moreover, coupled with EDS mapping and quantification (Figure 3b and Table S2) HRTEM confirms the presence of isolated iron-enriched areas. These areas predominantly show d-spacings of 2.00 ± 0.02 Å, 2.18 Å and 4.16 Å, all of which can account for the (131), (140) and (001) planes in goethite ($Fe_2O_3 \cdot H_2O$) (Table S2).[25] This observation suggests the formation of inhomogeneously distributed, small crystalline domains of goethite as a host material for the original iron species in addition to the iron incorporated in the $Ni(OH)_2$ structure.

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NiFe LDH shows very high catalytic activity for OER, with a current density of 10 mA cm^{-2} being reached at 0.34 V overpotential. Despite no significant structural changes were detected for the sample immersed in 1.0 M KOH at 25 °C for 60 h, the catalytic activity decreased compared to pristine NiFe LDH with an overpotential of 0.37 V being required to achieve 10 mA cm^{-2}. Additionally, whereas for pristine NiFe LDH the signals for OER and Ni^{2+} oxidation superimpose, a preoxidation shoulder is noticeable in the LSV of the sample immersed for 60 h in 1.0 M KOH. This observation results from a cathodic shift of the Ni^{2+} oxidation peak and the simultaneous anodic shift of the OER. A clear separation between Ni^{2+} oxidation and the OER was reported for $Ni(OH)_2$, where Fe incorporation (presence of Fe^{3+}) leads to an anodic shift of the Ni^{2+} oxidation, meanwhile the OER shifts to less anodic potentials.[8] Since the results obtained with other characterization methods did not suggest any major structural changes, it can be assumed that depletion of Fe from the NiFe layers only occurs to a small extent. This change nonetheless obviously influences the OER activity of the material, which is not surprising since it is known that the ratio between Ni and Fe is crucial for catalytic OER activity.[8,26,27]

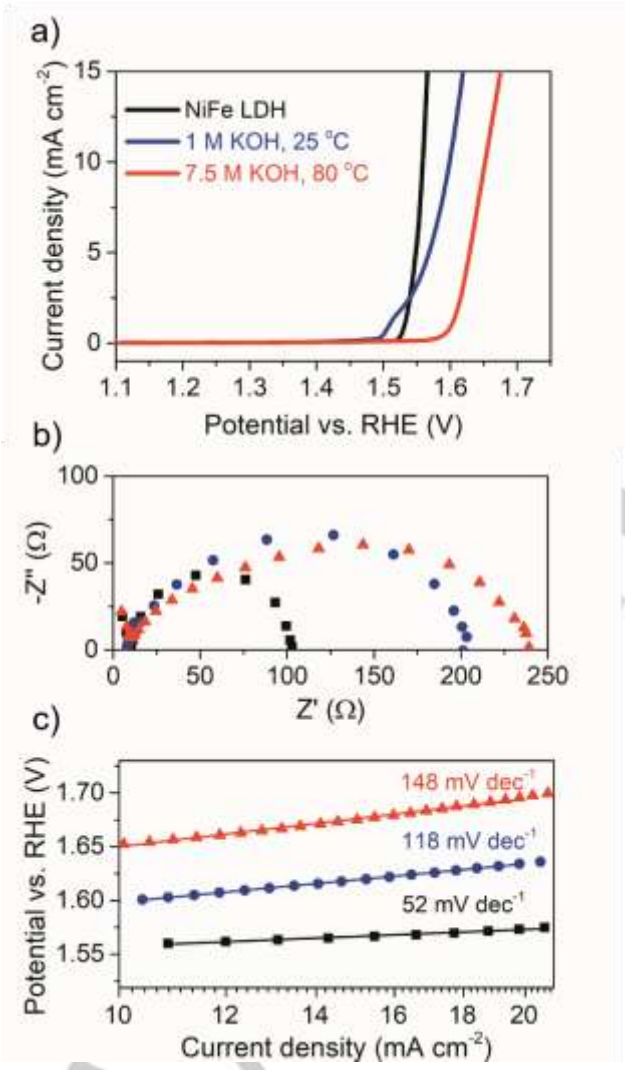

Figure 4. a) Linear sweep voltammograms registered at a scan rate of 5 mV/s and 1600 rpm, b) galvanostatic EIS recorded at 2 mA cm^{-2} in 1.0 M KOH and c) Tafel plots for NiFe LDH (black), and NiFe LDH after immersion for 60 h in 1.0 M KOH at 25 °C (blue) and 7.5 M KOH at 80 °C (red).

Moreover, a decrease in the slope of the LSV can be observed, indicating a decrease in the electrical conductivity of the material.[28] For the sample immersed in 7.5 M KOH at 80 °C, the LSV presented in Figure 4a shows a significant decrease in catalytic activity. A current density of 10 mA cm^{-2} is attained at an overpotential of 0.42 V, which is 90 mV higher than the overpotential at which pristine NiFe LDH delivers the same current density. This decrease in catalytic activity is evidently related to the formation of $Ni(OH)_2$ and FeOOH, both of which show lower catalytic activity than NiFe LDH.[26,28,29] Nyquist plots obtained from electrochemical impedance spectroscopy (EIS; Fig. 4c) at a similar current density, show a significant increase of the total charge transfer resistance for the samples immersed in KOH solution. This is attributed to the formation of FeOOH, which is known to have a low conductivity, leading concurrently to a diminished catalytic activity.[30] The Tafel slope for pristine NiFe LDH is 52 mV dec^{-1}, consistent with previously reported values.[10,28,31] For the samples immersed in 1.0 M KOH and 7.5 M KOH the Tafel slopes are 118 and 148 mV dec^{-1}, respectively, indicating slower reaction kinetics and prevalence of a different rate limiting step.[32]

In conclusion, chemical and structural transformation of NiFe LDH was investigated at conditions akin to industrial electrolysis, 7.5 M KOH (~30 % KOH) at 80 °C but without electrochemical polarization, as well in 1.0 M KOH at 25 °C and 80 °C. The immersion of NiFe LDH in 1.0 M KOH at 25 °C for 60 h instigated no significant changes on the structure of NiFe LDH, whereas its immersion in 7.5 M KOH at 80 °C for 60 h transformed the NiFe LDH into a mixture of crystalline β-$Ni(OH)_2$ and predominantly amorphous FeOOH with minor non-homogeneously distributed crystalline domains of α-FeOOH. For both samples aged either in 1.0 M at 25 °C or in 7.5 M KOH at 80 °C for 60 h, a substantial decrease in OER activity was observed. The decrease was significantly more drastic for the sample aged in 7.5 KOH. It is therefore recommended that the search and development of catalysts for OER must concurrently evaluate their stability at realistic conditions for industrial application.

Acknowledgements

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Keywords: NiFe LDH • chemical stability • electrocatalysis • oxygen evolution reaction • water splitting

[1] a) F. Song, X. Hu, *Nat. Commun.* **2014**, *5*, 4477; b) K. Fan, H. Chen, Y. Ji, H. Huang, P. M. Claesson, Q. Daniel, B. Philippe, H. Rensmo, F. Li, Y. Luo et al., *Nat. Commun.* **2016**, *7*, 11981; c) S. Barwe, C. Andronescu, J. Masa, W. Schuhmann, *Curr. Opin. Electrochem.* **2017**.

[2] F. Dionigi, P. Strasser, *Adv. Energy Mater.* **2016**, *6*, 1600621.

[3] B. M. Hunter, W. Hieringer, J. R. Winkler, H. B. Gray, A. M. Müller, *Energy Environ. Sci.* **2016**, *9*, 1734.

[4] Y. Xu, Y. Hao, G. Zhang, Z. Lu, S. Han, Y. Li, X. Sun, *RSC Adv.* **2015**, *5*, 55131.

[5] M. Gong, Y. Li, H. Wang, Y. Liang, J. Z. Wu, J. Zhou, J. Wang, T. Regier, F. Wei, H. Dai, *J. Am. Chem. Soc.* **2013**, *135*, 8452.

[6] a) S. Chen, J. Duan, M. Jaroniec, S. Z. Qiao, *Angew. Chem. Int. Ed.* **2013**, *52*, 13567; b) Q. Yang, T. Li, Z. Lu, X. Sun, J. Liu, *Nanoscale* **2014**, *6*, 11789; c) X. Lu, C. Zhao, *Nanoscale* **2015**, *6*, 6616; d) M. Gong, W. Zhou, M.-C. Tsai, J. Zhou, M. Guan, M.-C. Lin, B. Zhang, Y. Hu, D.-Y. Wang, J. Yang et al., *Nat. Commun.* **2014**, *5*, 4695.

[7] L. Trotochaud, S. L. Young, J. K. Ranney, S. W. Boettcher, *J. Am. Chem. Soc.* **2014**, *136*, 6744.

[8] M. W. Louie, A. T. Bell, *J. Am. Chem. Soc.* **2013**, *135*, 12329.

[9] S. Barwe, C. Andronescu, J. Masa, E. Ventosa, S. Klink, A. Genç, J. Arbiol, W. Schuhmann, *ChemSusChem* **2017**, *10*, 2653.

[10] C. Andronescu, S. Barwe, E. Ventosa, J. Masa, E. Vasile, B. Konkena, S. Möller, W. Schuhmann, *Angew. Chem. Int. Ed.* **2017**, *56*, 11258.

[11] K. Zeng, D. Zhang, *Prog. Energy Combust. Sci.* **2010**, *36*, 307.

[12] A. Mendiboure, R. Schöllhorn, *Rev. Chem. Miner.* **1986**, *23*, 819.

[13] D. G. Evans, R. C. T. Slade, *Structural Aspects of Layered Double Hydroxides*, In: X. Duan, D. G. Evans (eds) Layered Double Hydroxides. Structure and Bonding, vol. 119. Springer, Berlin, Heidelberg, **2005**.

[14] D. Tichit, M. N. Bennani, F. Figueras, J. R. Ruiz, *Langmuir* **1998**, *14*, 2086.

[15] R. W. Cairns, E. Ott, *J. Am. Chem. Soc.* **1933**, *55*, 527.

[16] T. N. Ramesh, R. S. Jayashree, P. V. Kamath, *Clays Clay Miner.* **2003**, *51*, 570.

[17] N. Han, F. Zhao, Y. Li, *J. Mater. Chem. A* **2015**, *3*, 16348.

[18] J. L. Bantignies, S. Deabate, A. Righi, S. Rols, P. Hermet, J. L. Sauvajol, F. Henn, *J. Phys. Chem. C* **2008**, *112*, 2193.

[19] D. S. Hall, D. J. Lockwood, S. Poirier, C. Bock, B. R. MacDougall, *J. Phys. Chem. A* **2012**, *116*, 6771.

[20] F. B. D. Saiah, B.-L. Su, N. Bettahar, *J. Hazard. Mater.* **2009**, *165*, 206.

[21] D. S. Hall, D. J. Lockwood, S. Poirier, C. Bock, B. R. MacDougall, *ACS Appl. Mater. Interf.* **2014**, *6*, 3141.

[22] M. Rajamathi, V. Kamath, R. Seshadri, *J. Mater. Chem.* **2000**, *10*, 503.

[23] P. Baraldi, G. Davolio, G. Fabbri, T. Manfredini, *Mater. Chem. Phys.* **1989**, *21*, 479.

[24] P. Delahay, M. Pourbaix, P. van Rysselberghe, *J. Chem. Educ.* **1950**, *27*, 683.

[25] G. Peacock, *Trans. R. Soc. Can.* **1942**, 116.

[26] D. Friebel, M. W. Louie, M. Bajdich, K. E. Sanwald, Y. Cai, A. M. Wise, M.-J. Cheng, D. Sokaras, T.-C. Weng, R. Alonso-Mori et al., *J. Am. Chem. Soc.* **2015**, *137*, 1305.

[27] D. A. Corrigan, *J. Electrochem. Soc.* **1987**, *134*, 377.

[28] X. Yu, M. Zhang, W. Yuan, G. Shi, *J. Mater. Chem. A* **2015**, *3*, 6921.

[29] a) O. Diaz-Morales, I. Ledezma-Yanez, M. T. M. Koper, F. Calle-Vallejo, *ACS Catal.* **2015**, *5*, 5380; b) M. S. Burke, M. G. Kast, L. Trotochaud, A. M. Smith, S. W. Boettcher, *J. Am. Chem. Soc.* **2015**, *137*, 3638.

[30] M. S. Burke, L. J. Enman, A. S. Batchellor, S. Zou, S. W. Boettcher, *Chem. Mater.* **2015**, *27*, 7549.

[31] Z. Lu, W. Xu, W. Zhu, Q. Yang, X. Lei, J. Liu, Y. Li, X. Sun, X. Duan, *Chem. Commun.* **2014**, *50*, 6479.

[32] T. Shinagawa, A. T. Garcia-Esparza, K. Takanabe, *Sci. Rep.* **2015**, *5*, 13801.

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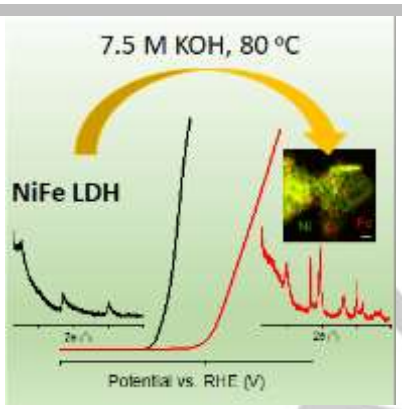
COMMUNICATION

Abundant non-noble metal catalysts exhibiting high stability and high catalytic activity towards the oxygen evolution reaction (OER) are required to improve alkaline water electrolysis. Laboratory tests usually are performed at gentle conditions as compared with those in industrial environment. Chemical transformation of NiFe LDH in high concentrated alkali solution at high temperature is reported and the impact on the catalytic OER activity is discussed.



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