International Journal of Science and Research (IJSR)
ISSN: 2319-7064
SJIF (2022): 7.942

Unlocking Oxygen Evolution: The Impact of Hetero Atom Doping on NiFe-Based Pre-Catalysts and Their Transformation into Ultrathin Layered Double Hydroxides

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Abstract: ***Green energy production by electrochemical water splitting is only able to fulfill the future global requirement for energy. Water molecules split into H_2 and O_2 by several electrocatalysts. The anodic oxidation of water for oxygen evolution reactions follows a kinetically sluggish mechanism and required some extra overpotential. In this regard, NiFe-based electrocatalysts are of low cost and highly efficient for OER activity. Herein, we have studied the effect of hetero-atom doping in the modulation of the electronic structure, increased number of active sites and improved electrochemical surface area enhanced the OER activity. The Nife-based electrocatalysts as pre catalyst are most renounced and transformed in to high valency active phase metal catalysts. In this work, we have proposed a mini review for the enhanced activity of hetero atom-doped NiFe-based Pre-Catalyst and their Structural Reconstruction into Ultrathin Layered Double Hydroxide for oxygen evolution reaction. The transformation in morphology and reconstruction of the surface in layered double hydroxide of NiFe-based pre-catalyst significantly enhanced the OER activity of the catalyst. The heteroatom doping enhances the synergic effect of the catalyst and enhanced electrochemical OER activity***

Keywords: heteroatom-doping; ultrathin nanosheets; electronic structure modulation; water splitting; oxygen evolution; catalysis

1. Introduction

The increasing population on earth there is greater demand of energy simultaneously environmental problems can be solved by green energy producing by electrochemical water splitting. The electrochemical water splitting is a great sustainable energy conversion technology [1,2]. The water splitting by electrochemical method results in to the to the production of H_2 and O_2. During water splitting two half-cell reactions were carried out such as hydrogen evolution reaction (HER) which occurs at cathode and oxygen evolution reaction (OER) which occurs at anode [3–5]. The water oxidation at anode is usually high energy process and required some extra overpotential of 1.23V vs RHE s it follow lethargic kinetics due to the transfer of four electron and proton mechanism [6–9]. Instead of this anodic OER involves formation of high energy intermediates [10–14]. This is the main cause by which extra energy as overpotential is required to overcome lethargic kinetics.

In this review we have studied some , noble metal based electro catalyst such as RuO_2, IrO_2 for electrochemical water oxidation which shows excellent catalytic activity [15–20]. But the large-scale production of these catalyst very costly because it involves rare earth natural abundance. There for it become very important to develop low cost electrocalyst with high natural abundance. Recently few year back first row transition metal based electrocatalyst has been developed as their oxides, chalcogenides, phosphides, carbide and nitrides has been explored due to their enhanced catalytic activity and stability [21–26].

In recent studies in is clearly exhibited the transition metal oxide, transition metal phosphide, transition metal chalcogenides and transition metal phosphide are good pre-electrocatalyst which transformed in to layered double hydroxide active catalyst under applied anodic potential in alkaline medium[4,13,21,27–30]. The cobalt phosphide (CoPn) nickel foam also shows great OER activity because it was electrochemically transformed in to layered double hydroxide under applied anodic potential and the cobalt phosphide catalyst showed oxygen evolution reaction only at 240 mV of overpotentials to achieve 10 $mAcm^{-2}$ current density[31].The formation of Co(O)OH active catalyst was established by in-situ XPS [14,32,33]. In the same way Hu et al developed NiFeP catalyst for electrochemical water splitting and this catalyst also readily converted in NiFeOOH active catalyst under applied anodic potential in alkaline medium. This showed excellent OER activity and achieve $10mAcm^{-2}$ current density only at overpotential of 219mV [5,16,34–38]. The reconstruction of pre catalyst in to active ultrathin layered double hydroxide possess high surface area , large number of exposed active sites, excellent electronic conductivity and improved charge and mass transfer which facilitates improvement of OER activity [6,11,35,39]. There are some other nickel and iron based cocatalyst such as NiOOH, FeOOH, NiFeOOH have been well explored for their excellent water splitting activity showing excellent oxygen evolution reaction [40–43]. In this regards we have explored some of the NiFe-based electrocatalyst and observed the effect of heteroatom doping.

Effect of hetero atom doping

The hetero-atom (such as S, N, P, etc.) doping boosts the catalytic activity of NiFe-based electrocatalyst and theses catalysts were showed excellent OER activity due to synergic effect of the catalyst (Fig-01). Some of the well renounced heteroatom doped electrocatalyst were as follows.

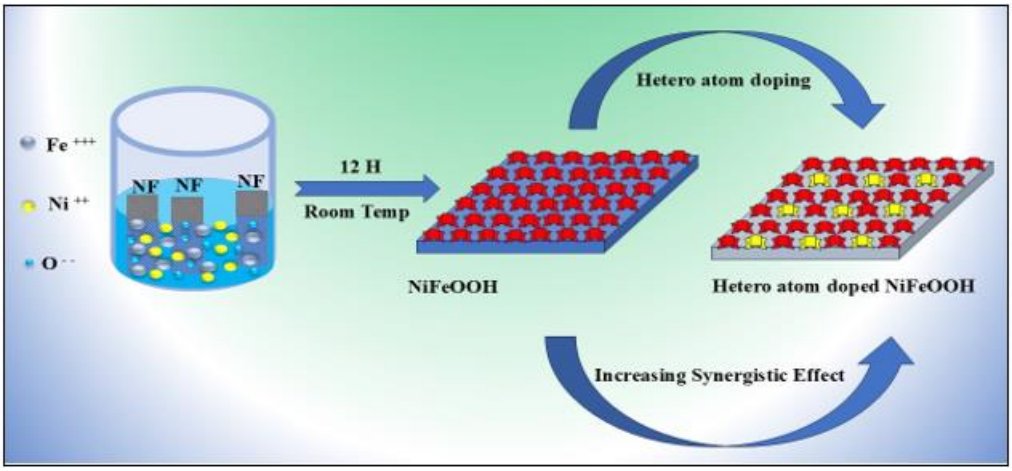

Figure 1: Schematic representation of preparation of NiFeOOH LDH and hetro-atom doping

a) Sulphur doped Electro-catalyst

Sulfur doping in Layered Double hydroxide (S-LDH) play important role in enhancing electro chemical water oxidation. This type of catalyst become a promising material for electrochemical water splitting and efficient catalyst for anodic water oxidation as well as hydrogen evolution reaction[44–46]. Herein when we look forward for the structural characteristics the Layered double hydroxides are a class of Layered materials which consists of positive charged layers of metal hydroxides, in between two positive layers negatively charged ions are present[47,48]. In case of Sulfur doped Layered double hydroxide (S-LDH) sulfur introduced in to interlayers site of positive metal ions. The electronic structure of LDH is modified by sulfur doping which enhance catalytic activity of S-LDH.

Sulphur atom doping greatly influences anodic water oxidation of water and show great OER activity therefore many compounds have been synthesized with Sulphur doping for electrochemical water splitting. The Sulphur doped FeOOH (S-FeOOH) electrocatalyst is utilized for excellent current output for oxygen evolution reaction which produces 1000 mA of current density with extremely low overpotentials of 358mV [49]. The catalyst S-FeOOH is industrial level electrocatalyst. This catalyst is very stable and produces 1000 mA cm current density for at least 1000h[43,50]. The enhanced rate of OER activity showed that Sulphur doping decreases energy barrier in rate determining step of oxygen evolution reaction. Hongli Wang et al prepared free-standing S-Ni/FeOOH@NF electrode which produces 10mA current density at only 229 mV of overpotentials in comparison NiFeOOH electrode having an overpotential of 409 mV to same current density[51,52]. The NiFeOOH catalyst is quite stable and remain 95% after 150 hours of its catalytic activity to produce 100 mA current density. In sulphur doped electrocatalyst enhanced OER activity due to high intrinsic catalytic activity and excellent hydrophilic nature. The Sulphur doping changes the electronic structure and charge density of NifeOOH electrocatalyst which optimize the OER activity in rate determining step which leads to the increase of binding energy of intermediate for OER activity which show enhanced catalytic activity. The Sulphur doped NifeOOH showed long term stability up to 200 hours with current density 399 to 500 mA cm so it is suitable for industrial level electrocatalyst[53].

b) Nitrogen doped Electro-catalyst

Nitrogen is another important hetero atom which is used extensively as dopant in transition metal based electrocatalyst to improve OER activity of electro catalyst. Huge amount of hydrogen energy can be produced by utilizing sea water which is much sustainable and promising method without destroying pure water resources[54–56]. Thus, sea water electrolysis is most prominent route to produce energy but only obstacle is the chlorine oxidation reaction occurring at anode which produces highly toxic chlorine species as well as corrodes anode[57]. The Chlorine oxidation reaction were well suppressed by Nitrogen doped carbon dots strongly coupled NiFe layered double hydroxide nanosheet on Ni-Foam which is utilized for oxygen evolution reaction (OER) at extremely low over potential of 260 mV with producing 100 mA cm^{-2} of current density in alkaline water and this electrode is very suitable for oxidation of sea water[57]. Fabricated Nife layered double hydroxide at nitrogen doped TiO_2 nanotube (NiFe/N-TiO_2) is another important electrocatalyst and well utilized for oxidation evolution reaction. In NiFe/N-TiO_2 only 20.9% nitrogen dopant showed enhanced OER activity at very low over potential at 235 mV to achieve 10 mA cm^{-2} of current density having very low slop of 48.9 mV dec^{-1} in Tafel plot and this catalyst is highly stable[56]. The nitrogen doped TiO_2 exhibited large surface area having nanotube structure. Nitrogen doping enhances the electrical conduction as well as weakens the oxygen interaction on NiFe-LDHs catalyst. The development of mesoporous N-doped LDHs is cost effective and enhanced OER as well as HER activity.

c) Phosphorus Doped Electro-catalyst

The phosphorous doped LDHs is another most efficient technique for cost effective high performance & highly stable bifunctional electrocatalyst to replace noble metal catalyst for enhanced over all water splitting to produce green and renewable hydrogen energy along with anodic oxidation produces clean oxygen[58]. The bi-functional NiFe phosphite have been developed on NiFe alloy foam which exhibited 3D coral-like morphology in alkaline medium. This coral-like structure leads to large exposer active sites of catalyst which showed enhanced catalytic activity due to huge charge transport and bubble released. The bicomponent of catalyst NiFe phosphite exhibited synergistic effect which leads to strong interaction between different metal active sites and showed excellent HER only at an over potential of 132 mV at

10 mAcm^{-2} of current density and OER only at overpotentials of 181 mV 10 mA cm^{-2} current density. The phosphorus doped NiFe alloy nano particles at carbon cloths (P-Ni0.5fe@C) synthesized by hydrothermal method also cost effective non precious catalyst exhibited hollow structure and excellent adjustment of phosphorus dopant with carbon encapsulated active component provides large number of active sites for enhanced water splitting activity. P-Ni0.5fe@C showed OER activity at very low overpotential of 256 mV to achieve 10 mA cm^{-2} current density and very small Tafel slope of 65 mV dec^{-1}, this catalyst also performed excellent long-term stability towards OER activity in 1M KOH which superior than IrO_2 electrocatalyst showing 318 mV over potentials to achieve 10 mA cm^{-2} current density.

The black phosphorus is known for good electrocatalyst for OER activity but low stability and poor intrinsic activity is major problem but when NiFe-LDH is assembled on the exfoliated surface of black phosphorus (NiFe-LDH@EBP) gives out nanoflakes structure. The nanoflake structure facilitate the interfacial coupling to achieve enhanced electrocatalytic activity and good stability. In NiFe-LDH@EBP interfacial P-O bonding showed high OER activity at only overpotential of 240 mV to achieve 10 mA cm^{-2}. DFT calculation support the interfacial interface coupling of NiFe-LDH of black phosphorus promotes the charge transfer kinetics which improves catalytic activity.

Another Phosphorus doped NiFe electrocatalyst is P-$Ni_{0.75}Fe_{0.25}Se_2$. This electrocatalyst performed superior electrochemical catalytic activity towards oxygen evolution reaction and achieved 100mA cm^{-2} current density only at 238 mV of overpotentials which is much better than RuO_2 and $Ni_{0.75}Fe_{0.25}Se_2$ electrocatalyst. P-$Ni_{0.75}Fe_{0.25}Se_2$ catalyst is highly stable in alkaline medium.

The Phosphorus doped NiMo catalyst is another significant example of showing effect of Phosphorus doping in electro catalytic activity. The P-doped NiMo bimetal aerogel is cost effective electrocatalyst for electrochemical water splitting. P-doped NiMo electrocatalyst show extremely low over potential of 69 mV for hydrogen evolution reaction and 235 mV overpotential for oxygen evolution reaction to achieve 10mA cm^{-2} current density. The excellent HER and OER activity of P-doped NiMo is due to redistribution of electron Mo alloying and P-doping.

2. Conclusions and Perspectives

In this paper we have concluded that NiFe based catalyst show promising catalytic activity for electrochemical water splitting and can be best alternative of noble metal catalyst. The noble metal catalysts are of costly in comparison to NiFe-based materials. The NiFe based catalyst cost effective and cheap catalyst having great catalytic activity large number of active phases and flexible compositional variation. During catalytic activity these catalysts were in situ converted in to high valent state along with tuning in composition. The NiFe based pre-catalyst are easily transformed in to Layered doble hydroxide which is active catalyst for electrochemical water splitting. The careful development of catalyst, by careful tunning of morphology, surface structure and chemical environment one can prepare improved and enhanced OER electrocatalyst. The surface structure modifications by doping appropriate elements, composition modification, tunning chemical state and heterogeneous constitution leads to production of cost effective excellent electrocatalyst with superior electrochemical water splitting activity. The structural and electronic modifications facilitate the adsorption of intermediates, charge transfer, synergistic effect and metal (oxy)hydroxide active catalyst formation. The hetero atom doping effects results in to the increase of current density at lower applied potential at RHE in comparison to undoped electro-catalyst (Fig-02). Thus hetero-atom doped NiFe based catalyst express great hope in future electrochemical water splitting catalysis.

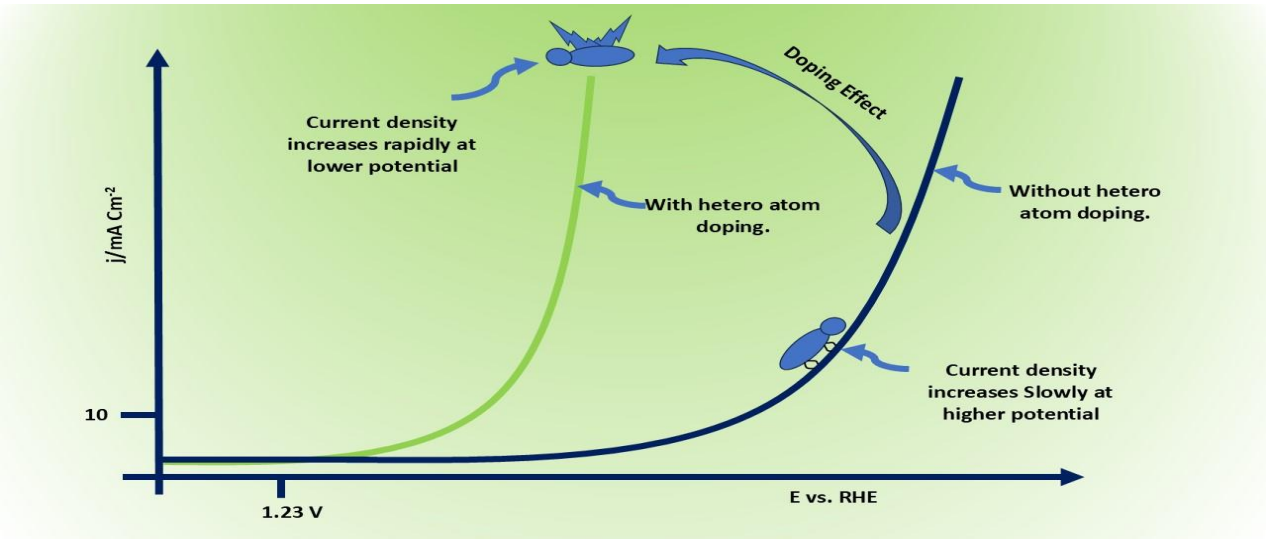

Figure 2: Schematic representation of variation of current density in NiFeOOH LDH and hetero-atom doped X-NiFeOOH LDH against applied potential at RHE.

Conflicts of interest

"There are no conflicts to declare."

Author Contribution

This review article is written by Manoj Kumar under the supervision of prof (Dr) Raghwendra Pratap Singh

K.N.I.P.S.S. Sultanpur U.P India. The review article is written by the contribution of all co-authors. All the authors have given the approval to the final version of manuscript. The conceptualization, editing and reviewing was performed by Dr. R. P. Singh**.**

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