Pemanfaatan Air Sebagai Sumber Blue Energy untuk Produksi Gas Hidrogen dengan Desain dan Fabrikasi Katalis yang Efisien

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Abstract

Creating an efficient electrocatalytic hydrogen evolution reaction (HER) by developing stable and earth-abundant materials is an essential and highly efficient method to meet the increasing demand for sustainable energy. For this reason, we have proposed a simple and novel Covalent triazine frameworks (CTFs) composite with Fe and Ni as catalysts for highly active electrocatalysts (ECs) HER in a typical alkaline electrolyte. The detailed chemical and morphological characterizations were performed by FT-IR, SEM-EDX and XRD. The as-synthesized CTF-Ni ECs exhibited efficient and stable HER performances with low overpotentials of 260 mV to attain a 10mA cm^{-2} current density. The addition of metal improves the electron transfer and creates active sites of water dissociation and combining hydrogen intermediates to hydrogen gas (H_2) sites in HER. These results show that a simple novel synthesis approach for the metal-hybrid CTFs could be an alternative to using noble metals for sustainable energy-related applications.

Keyword: Keywords: Covalent triazine frameworks, electrocatalysis, hydrogen evolution reaction, water splitting

Introduction

Covalent triazine frameworks (CTFs) are carbon-based materials with potential for use as electrocatalysts in the hydrogen evolution reaction (HER). These materials are composed of triazine or tri-s-triazine structural units linked by tertiary amines. CTFs have good chemical and thermal stability, strong electronic density in material surfaces, and energy-gap tunability. While they possess limited electronic conductivity, The performance of CTFs can be further enhanced through modification with foreign atom doping. Noble metal doping is known improve CTFs performance in electrocatalyst applications. However, the scarcity of noble metals hindered the large-scale applicability of CTFs. Thus, less noble metals such as transition metals, which is way more abundant could be used as noble metals alternative as dopant in CTFs electrocatalyst applications. Transition metals, such as Fe, Ni, Mn, and Co have been studied for electrocatalytic applications However, to improve performance and stability of those metals, the composite of CTFs with high stability and transition metals with better electronic conductivity could be studied.

Research Method

a. Materials and Reagents

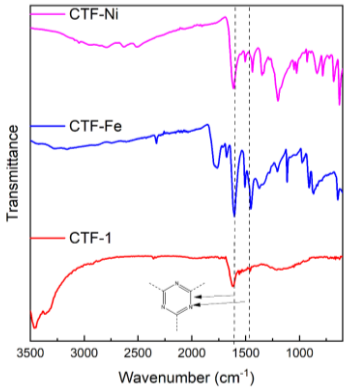
Reagent-grade chemicals were obtained from various suppliers. 1,4 Dicyanobenzene (DCB) (CAS No. 623-26-7, Sigma Aldrich, Germany) was used as the precursor for CTF synthesis, while $ZnCl_2$ (CAS No. 7646-85-7, Supelco, U.S) served as both the ionic melt medium and reaction facilitator. $FeCl_3$ (CAS No. 7705-08-0, Sigma Aldrich, Germany) and $NiCl_2$ (CAS No. 7718-54-9, Sigma Aldrich, Germany) was introduced as source of metal doping. KOH (CAS No. 1310-58-3, SupelCo, U.S), Nafion® 117 solution (5 %, Sigma-Aldrich), absolute ethanol (EtOH, ≥99.9 %, Chemsolute), HCl (CAS No. 7647-01-0, SupelCo, U.S), and deionized (DI) water were used. All materials were used as received from the manufacturers without any further purification.

b. CTF-Fe and Ni catalyst preparation

Three CTFs catalyst denoted as CTF-Fe and Ni was prepared via a modified ionothermal method. DCB (0.5 g) was mixed with a $ZnCl_2$ (2.25 g) and three different ionic salts, $FeCl_3$; and $NiCl_2$ (0.5 g) respectively. The mixture was then heated in sealed ampoule at 350°C for 24 h in a furnace. The resulting product was washed repeatedly with HCl, ethanol, and DI water. Finally, the ingredients were dried in an oven at 90°C for 12 h.

c. Electrocatalyst studies

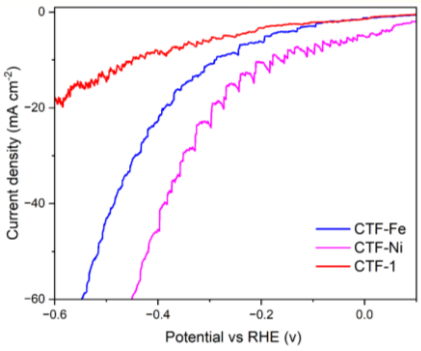
All electrocatalytic measurements of the three-electrode systems were performed using a potentiostat/galvanostat controlled by the DropView 8400 software. The three-electrode measurement setup consisted of Ag/AgCl (3 M KCl) as the reference electrode (RE), Pt mesh as the counter electrode CE, and carbon paper (CP) as the working electrode (WE). The potential (E) of the three-electrode systems was converted to the reversible hydrogen electrode (RHE) scale based on the following equation: E [V vs. RHE] = E(Ag/AgCl) + 0.2171 + 0.059 × pH. Linear sweep voltammetry (LSV) polarization curves were acquired using a 1 M KOH solution at a scanning rate of 5 mV/s. The scanning ranges for the OER and HER were 0–1.1 V and 1.2–1.8 V vs. RHE, respectively. Cyclic voltammetry (CV) curves were recorded by the scanning rate from 20 mV/s with a step size of 20 mV.



Picture 1. FT-IR Spectra of the Catalysts

Discussion & Result

$ZnCl_2$ as ionothermal molten salt medium. The Fourier-transform infrared (FT-IR) of the all of the CTFs materials, CTF-1; CTF-Fe and CTF-Ni showed prominent peaks at 1590 cm^{-1} for -C=N stretching nodes and 1420 cm^{-1} for C-N stretching nodes [13]. CTF-Fe, however, has additional peaks at 1700 cm^{-1} for carbonyl (C=O) groups. Also, ~1200 cm^{-1} peak as well, which can be attributed for C-O stretch. the low wavenumber peak of CTF-Fe could be described for M-O or Fe-O vibrational bond [14]. CTF-Ni has the same strong peak with CTF-Fe at ~1200 cm^{-1} for C-O stretch. The metal coordination of CTF-Ni could be ascribed by strong low wavenumber peak at ~500-400 cm^{-1} for M-N or N-Ni bonds [13, 15]



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The electrocatalytic HER activity of all CTFs materials in 1 M KOH was evaluated via linear sweep voltammetry (LSV). As shown in fig 4, the obtained CTF-1 has required overpotentials (η) exceeding 400 mV to achieve current density of -10 mA cm^{-2}, which reveals poor catalytic activity. The addition of metals, Fe and Ni in the CTFs synthetic by adding its metal salt, namely $FeCl_3$ and $NiCl_2$, improves the catalytic performance of the CTFs materials. CTF-Ni requires overpotentials of around 260 mV and CTF-Fe is 311 mV to achieve the same current density.

Conclusion

CTF-1 material was synthesized using well-known ionothermal methods. By further modified by adding another transitional metal ionic salts such as $FeCl_3$ and $NiCl_2$ to the ionothermal, the CTF-Ni and CTF-Fe were successfully synthesized along with CTF-1. CTF-Fe and CTF Ni were composed of hematite α-Fe_2O_3 material and α-$Ni(OH)_2$ and β-$Ni(OH)_2$ respectively. The HER studies of the CTFs electrocatalyst demonstrated that metal addition significantly enhances the HER performance. Among the tested catalyst, CTF-Ni has superior performance followed by the CTF-Fe and pristine CTF-1. The addition of metal improves the electron transfer, and the metal also acts as active sites of water dissociation and combining hydrogen intermediates to hydrogen gas (H_2) sites in HER.