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# Monolithic nanoporous Ni–Fe alloy by dealloying laser processed Ni–Fe–Al as electrocatalyst toward oxygen evolution reaction

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## ABSTRACT

In this paper, monolithic nanoporous Ni–Fe electrocatalyst is developed by dealloying a Ni–Fe–Al alloy, fabricated by a laser-based manufacturing method, as the electrocatalyst toward oxygen evolution reaction (OER). The nanoporous Ni–Fe alloy displays an improved electrochemical performance by exhibiting an OER current density of 100 mA/cm<sup>2</sup> at 442 mV overpotential in the 1 M KOH aqueous solution, which is better than that of laser processed bulk Ni–Fe alloy (464 mV). The structures, crystallinities, and chemical compositions of the nanoporous Ni–Fe electrocatalyst are characterized by SEM, XRD and EDXS. With an activity comparable to the electrocatalysts reported to date, the monolithic nanoporous Ni–Fe electrodes are also highly flexible in mechanical configurations, due to the combined advantages of both laser and dealloying processes, which provide new options for OER electrode design.

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## Background

Since clean energy and environmental protection attract global concerns, the demand for renewable and clean energy sources has been increasing rapidly [1–9]. Water electrolysis is a promising method for hydrogen generation [10–14]. Oxygen evolution reaction (OER) is the redox reaction occurring at the anode and electrolyte interface during water electrolysis, which is also the sluggish portion of water splitting. The reaction rates strongly depend on the applied overpotential. With a suitable heterogeneous electrocatalysis, due to the formation of intermediate species, an energy shortcut at a much lower overpotential is allowed. However, due to the general needs of expensive noble metal-based

electrocatalysts, such as Ir, Ru, and Pt, the commercialization of water splitting is hindered [15–17]. Recently, more research efforts have been devoted to explore earth-abundant and eco-friendly electrocatalysts to replace noble metals. The reported materials for OER [18–21] include transition metals and alloys [22–25], metal oxides [26–28], metal chalcogenides [29], metal pnictides [30,31], organometallic [32] and non-metallic materials [33–35].

The most decisive factor for the catalytic activity of an electrocatalyst is the intrinsic binding strength toward the reaction species, though further modifications could improve the performance to some degree. A moderate strength is required for an electrocatalyst as stronger bindings would hinder product repelling while weaker bindings would result in difficult intermediates stabilization [36]. It is found that d-

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block metals usually have a moderate absorption to main group elements; since there are unpaired electrons on their outermost shell s orbitals and secondary outermost shell d orbitals [37,38]. In addition, a proper geometric configuration of the electrocatalyst would strongly increase the catalytic activity. If the lattice spacing is too large, diatomic reactant absorption and diatomic product formation would be inhibited; and if atoms are too closely packed, strong reactant repulsion would be resulted [39,40]. Recently, Ni–Fe alloy stands out as a most appealing category for OER electrocatalysts, due to its moderate absorption affinity to reaction intermediates, good match with the reaction coordination, and low material cost [41–45]. Along with an ideal composition, an optimal electrocatalyst also requires a high specific surface area, which not only influences the localized electronic structure of surface atoms, but also alters the availability of active sites [46–49]. Nanostructuring is the most straight forward way to modify the structure of a catalyst, owing to the minimization of particle sizes and the increased exposed specific surface area. Therefore a nanostructured Ni–Fe alloy possesses a great potential for low-cost and high-performance OER electrocatalyst.

Compared to traditional powder-based electrocatalysts, monolithic electrocatalyst displays great advantages in simplifying the electrode assembling process and avoiding electrical connection issues [50]. For example, for hydrogen generation, Xie et al. reported a self-standing monolithic cobalt oxide array for  $\text{NaBH}_4$  hydrolysis that effectively inhibited catalyst aggregation and avoided the tedious and time-consuming process for catalyst separation [51]. As a rapidly

developing manufacturing method, laser-based additive manufacturing is an excellent technique to prepare complex templates for monolithic electrocatalyst. In addition, laser-based additive manufacturing can achieve the desired alloy compositions without tedious chemical synthesis and can achieve a strong metallic bond to the substrate avoiding the usage of binders. For instance, Laser Powder Stream, or LENS<sup>®</sup>-Laser Engineered Net Shaping<sup>™</sup> process is advantaged in free forming/coating capability to make large and complex parts. Due to the small laser spot size and thus a rapid cooling rate, laser processed parts have unique microstructures and material properties. For example, Cebollero et al. developed thin ceramic membranes for electrolyte-supported solid oxide fuel cells through laser processing which effectively improved the electrolyte-electrode contact and thus reduced cathodic polarizations [52]. Dealloying is an efficient strategy to develop nanostructures, which can develop microscale channels to facilitate mass transfer and nanoscale features to provide more active sites at the same time [53–55]. In this regard, Xu et al. fabricated nanoporous PtCo and PtNi alloy ribbons through a mild dealloying process. Those materials displayed excellent activity and stability toward methanol electro-oxidation [56]. Herein, monolithic nanoporous Ni–Fe alloy electrodes are developed by dealloying laser-processed Ni–Fe–Al alloy samples as the electrocatalyst toward OER. Its crystal structures, morphologies and chemical compositions are characterized by XRD, SEM and EDXS. Its electrochemical performance toward OER is evaluated by linear sweep voltammetry, Tafel slope, and electrochemical impedance spectrometry.

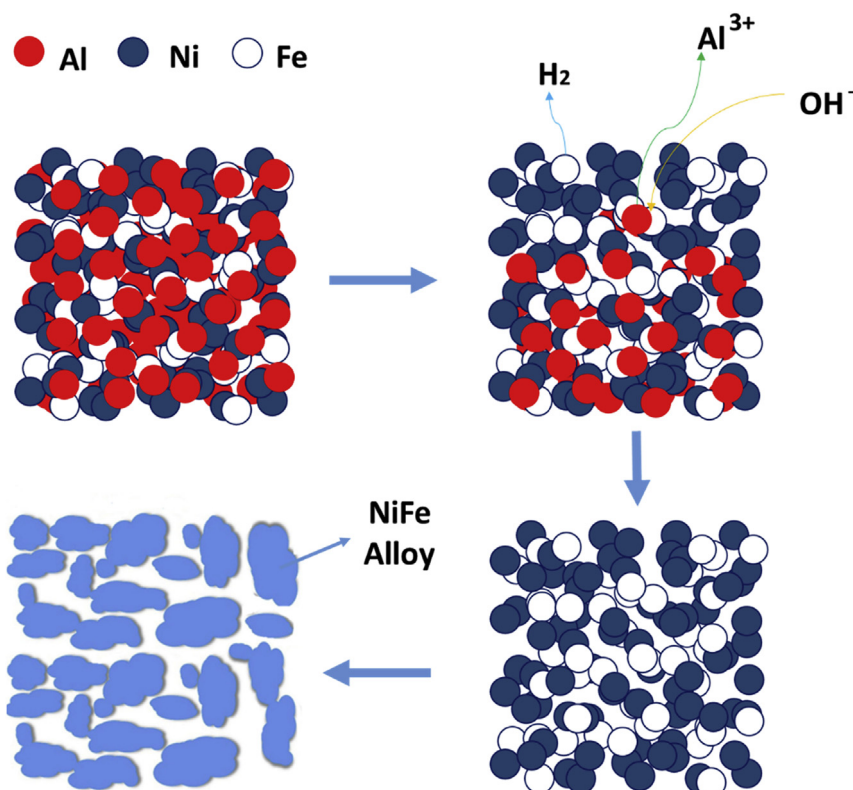


Fig. 1 – Illustration of Ni–Fe–Al dealloying process.

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