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# Cold spray as a novel method for development of nickel electrode coatings for hydrogen production

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## ABSTRACT

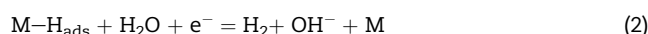
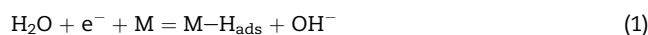
Cold spraying process was employed as a novel method to prepare electrocatalysts for hydrogen evolution by the water electrolysis process. Three electrode coatings were developed by cold spraying of nickel powder on aluminum substrates employing different coating parameters. The effect of the electrodes surface topography, microstructure, residual stress and surface strain on the electrocatalytic activity of the coatings was investigated using steady state polarization curves and electrochemical impedance spectroscopy. The results revealed that the largest electrocatalytic activity was obtained for the electrode with the lowest deposition efficiency that experienced the highest strain accumulation and compressive residual stress. It is expected that the large densities of dislocations along with a more favorable electronic structure of this coating caused by the peening effect encountered by the bounced-off particles were responsible for the higher activity of this electrode.

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## Introduction

High purity hydrogen produced by alkaline water electrolysis can be used as a green and renewable energy resource with no emissions other than water vapor when used as a fuel [1,2]. In a basic water electrolysis process, a direct current passes through cathode and anode electrodes placed in a conductive electrolytic solution and decomposes water into hydrogen and oxygen [1]. Hydrogen evolution reaction (HER) starts with the discharge of a water molecule (Volmer step, Eq. (1)) followed by either electrochemical desorption (Heyrovsky step, Eq. (2))

or chemical recombination (Tafel step, Eq. (3)) depending on the hydrogen–metal bond strength:



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High energy consumption of the electrolysis process has made large scale hydrogen production by this method economically unattractive [1–4]. However, the energy efficiency of water electrolysis can be improved by reducing the hydrogen and oxygen evolution overpotentials that contribute significantly to cell energy losses [3–6].

Using highly intrinsically active electrode materials reduces the internal voltage losses and increases the efficiency of the HER [6–9]. Nickel is one of them and is widely used as the electrode material for alkaline water electrolysis due to its high electrocatalytic activity, availability, stability in alkaline solutions, and relatively low cost [8–12]. Apart from material selection, the physical morphology of an electrode has also a significant influence on the overpotential, and thus, on its electrocatalytic performance. A larger effective electrode surface adds to the reaction area by providing more electrode-electrolyte interface, reducing the operating current density, and contributing to a higher electrocatalytic activity [9,13,14]. The surface area can be enhanced by increasing the electrode surface asperities, such as surface roughness and porosity.

Furthermore, surface crystallography of an electrode substantially affects its electrocatalytic behavior. Enhanced performance of nanocrystalline catalysts for the HER as a result of high volume fraction of surface defects has been extensively discussed in the literature [15–20]. Improved electrocatalytic activities of nanostructured materials could be attributed to their favorable electronic structure generated due to the increased amounts of atoms located at interfaces between adjacent grains, which are likely to be out of equilibrium [18,20]. These surface properties depend strongly on the technique used to prepare the electrode surface. The possibility to deposit coatings with minimal oxidation and ultrafine microstructures due to severe plastic deformation makes cold spraying a promising technique for producing active electrode coatings for electrochemical processes in general and alkaline water electrolysis in particular.

Cold spray, invented in the mid-1980s, has been the object of intensive development over the last 20 years. This thermal spraying technique permits deposition of coatings in an all-solid-state process [21,22]. In a cold spray process, solid powder particles (typically 1–50  $\mu\text{m}$  in diameter) are accelerated toward a substrate at high velocities by a supersonic jet of compressed gas that has been cooled down to temperatures close to room temperature, well below their melting point. The compressed gas is generally nitrogen, air, or helium flowing through a converging-diverging nozzle. Consequently, upon impact, the particles experience plastic deformation and shear instability at the interface with the substrate or the already deposited layers. This also causes disruption and removal of the oxide films from the surfaces of both substrate and particles, and promotes the bonding by providing an intimate contact between them. Finally, they mechanically bond to the substrate and form a three-dimensional coating having a good bond and cohesion strength [21,23–25]. For the particles to deposit on the substrate, their velocity upon impact needs to be higher than a so-called critical velocity, which depends on the material, size and morphology of the particles. Generally, the particle impact velocity increases by increasing the temperature and pressure of the propellant gas and by reducing the particle diameter [22,26–29].

Since the gas temperature decreases rapidly upon acceleration, the temperature of the spray particle stays relatively constant and neither oxidation nor phase transformation occur in the in-flight particles [26,30]. As-sprayed coatings are generally in compression due to the impingement of the spraying particles on the surface at high velocities inducing peening effect [30]. A schematic view of a cold spray system is depicted in Fig. 1.

The present study investigates the possibility of developing electrocatalytically active electrode coatings by cold spray and studies the electrochemical properties of the produced electrodes. The effect of cold spraying parameters on microstructure and electrochemical activity of the developed electrodes was studied. Steady state polarization curves and electrochemical impedance spectroscopy (EIS) measurements were used to examine the electrocatalytic activity of the coated nickel electrodes towards HER. The best coating conditions were determined for development of the electrode with the highest electrocatalytic activity.

## Material and methods

### Cold spray process

Commercially available gas atomized nickel powder (Metco 56C-NS) was used as feedstock powder to produce cathode electrode coatings on aluminum (Al 6061) substrates. The particle size distribution of the powder was measured with a laser diffraction particle analyzer (Malvern Instrument's Spraytec) using a 300 mm lens. Fig. 2 illustrates the measured particle size distribution that ranges from 45 to 95  $\mu\text{m}$ . The distribution widths of D10, D50, and D90 were measured to be 54, 66, and 78  $\mu\text{m}$ , respectively.

The as-received morphology and cross sectional SEM images of the atomized nickel powder are shown in Fig. 3a and b, respectively. As shown in the figures, the powder particles were spherical with a dense structure containing nearly no porosity.

Table 1 provided by the powder manufacturer shows the nominal chemical composition of the powder determined using an Inductively Coupled Plasma (ICP) system.

To study the effect of the temperature and pressure of the propellant gas on the structure and electrocatalytic activity of the electrodes, three cold sprayed samples were deposited according to the spray conditions displayed in Table 2. Coatings were produced by Plasma Giken (Webster Massachusetts, USA) with their Model PCS-1000 cold spray system. In all experiments, two passes were deposited using nitrogen as the propellant gas, at 25 mm standoff distance, 50 g/min powder feeding rate, and 300 mm/s gun traverse speed.

### Coating characterization

For metallographic studies, as-sprayed samples were sectioned perpendicular to the coating surface using a low speed precision cut-off machine and then cold mounted. Subsequently, the samples were ground and polished according to the conventional metallographic procedures up to 0.05  $\mu\text{m}$  diamond suspension surface finish. The cross section

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