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### **ACCEPTED MANUSCRIPT**

## Structural and electronic optimization of graphene encapsulating

### binary metal for highly efficient water oxidation

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#### Abstract

Encapsulating non-precious metals within graphene layers represents a promising strategy to substitute precious metal catalyst towards the oxygen evolution reaction (OER). The surface electronic structure of graphene can significantly affect the OER performance, which depends on the types of encapsulated metal and their proportion but it still lacks efficient methods to modulate them. Herein, we report a universal strategy to encapsulate FeNi binary metal nanoalloy within ultrathin graphene layers, which can efficiently optimize the electronic properties and the OER activity on the graphene surface via modulating Fe/Ni ratio. The optimized catalyst with Fe/Ni of 1 shows a low overpotential of 280 mV at the current density of 10 mA cm<sup>-2</sup>. Both the catalytic activity and durability of the catalyst are better than the commercial IrO<sub>2</sub>. Theoretical calculations indicate that the adsorption strength of each intermediate on graphene can be optimally balanced by modulating the metal proportion of the encapsulated FeNi, leading to an enhanced OER activity with reduced overpotential on the graphene surface.

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