

# Accepted Manuscript

Tuning active sites on cobalt/nitrogen doped graphene for electrocatalytic hydrogen and oxygen evolution

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## Responses to Reviewers

We express our special thanks to the reviewers since your comments have guided us to improve the quality of the manuscript. Based on the comments, the authors have revised the paper accordingly. The changes have been marked in red color in the revised manuscript. The detailed responses have been listed below.

### REVIEWER 1 COMMENTS

This manuscript described a systematic study of the role that Co-NG catalysts have on the HER and OER activities. It was found that a well-controlled geometric formation of active Co-N and Co-C sites is the key factor for enhancing HER activity. The authors also found that Co-O chemical bonding is the key role for OER activity. The results of this work suggest new ideas of the electrocatalytic field. However, I would support the publication of this manuscript only after the authors address the following issues:

1. Introduction: The introduction part seems not demonstrating enough motivation of the study. In order to meet the high publishing standard of *Electrochimica Acta*, please revise the last paragraph of the introduction part to clearly illustrate the research motivation and highlights.

Response: Thank you for your good suggestion. We have revised last paragraph of the introduction part to clearly illustrate the research motivation and highlights. The related contents have been added in revised manuscript as bellow:

Since the distribution/aggregation and surface bond states of cobalt atoms on those carbon materials are diverse and the according explanations of catalytic activity have been totally distinct, which make the understanding of catalytic origin and material design direction in different ways.<sup>34</sup>

Motivated by great value of understanding origin of HER and especially OER activities for cobalt/carbon based composites, we have systematically investigated aggregation and bond states of cobalt atoms on basal plane of nitrogen doped graphene via synthesized a series of cobalt/nitrogen doped graphene (Co-NG) composites. The controlled dispersion of cobalt atoms on graphene basal plane and their bond state evolution from Co-O bonds to Co-C and Co-N bonds were achieved via tuning synthetic temperature, mass ratio of precursors, and structure of GO. Atomically dispersed cobalt atoms with enhanced Co-N/Co-C bonds on defect controlled graphene

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