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## **ACCEPTED MANUSCRIPT**

# Atomically dispersed Au<sub>1</sub> catalyst towards efficient electrochemical synthesis of ammonia

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#### Abstract

Tremendous efforts have been devoted to explore energy-efficient strategies of ammonia synthesis to replace Haber-Bosch process which accounts for 1.4% of the annual energy consumption. In this study, atomically dispersed  $Au_1$  catalyst is synthesized and applied in electrochemical synthesis of ammonia under ambient conditions. A high  $NH_4^+$  Faradaic efficiency of 11.1% achieved by our  $Au_1$  catalyst surpasses most of reported catalysts under comparable conditions. Benefiting from efficient atom utilization, an  $NH_4^+$  yield rate of 1,305  $\mu$ g h<sup>-1</sup>  $mg_{Au}^{-1}$  has been reached, which is roughly 22.5 times as high as that by supported Au nanoparticles. We also demonstrate that by employing our  $Au_1$  catalyst,  $NH_4^+$  can be electrochemically produced directly from  $N_2$  and  $H_2$  with an energy utilization rate of 4.02 mmol  $kJ^{-1}$ . Our study provides a possibility of replacing the Haber-Bosch process with environmentally benign and energy-efficient electrochemical strategies.

#### **Keywords**

NH<sub>3</sub> synthesis, metal single sites, electrocatalysis, Haber–Bosch process, nitrogen reduction

#### 1. Introduction

As one of the most essential industrial chemicals, ammonia (NH<sub>3</sub>) is currently produced on an enormous scale of over 150 megatons per year by the Haber–Bosch process which requires pressures of 200 to 300 atmospheres and temperatures from 300 to 500  $^{\circ}$ C. To date, this energy-and capital-intensive process accounts for 1.4% of the annual energy consumption and around 3% of global CO<sub>2</sub> emissions [1-5]. Electrocatalytic approach, especially driven by renewable energy,

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