Precise Synthesis of Nickel Sub-nano Particles for Greenhouse Gas Conversion

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Sub-nano particles (SNPs) have attracted worldwide attention due to the high reactivity and the unique property beyond the extension of the bulk and nanoparticles. Our research group has achieved the precise synthesis of various SNPs showing the higher catalytic performance than that of conventional materials by the template method using a dendrimer.[1,2] In this work, we succeeded to synthesize 4 types of nickel SNPs and applied them to greenhouse gas conversion as catalysts.

Following accumulation of nickel salts to dendrimers, 4 types of nickel SNPs were prepared by reduction of these complexes. The dendrimer applied in this work possesses intramolecular potential gradient, thereby the precise synthesis of nickel SNPs with defined atomicity such as Ni_4, Ni_{12}, Ni_{28}, and Ni_{60} was realized. (Fig. 1a) This stepwise complexation behavior was monitored by UV-vis absorption spectroscopy. STEM observation and EDX analysis demonstrated that SNPs with a diameter of around 1 nm composed of nickel atoms were loaded on silica and graphene supports. (Fig. 1b) The oxidation state and H_2-reduction temperature of nickel SNPs were evaluated by XPS, H_2-TPR, and XAFS measurements. Remarkably, it was indicated that nickel SNPs had the higher catalytic performance than conventional materials and the outstandingly durability for the greenhouse gas conversion reaction. It was theoretically suggested that this result originated from the structure peculiar to SNP with the unique active sites.

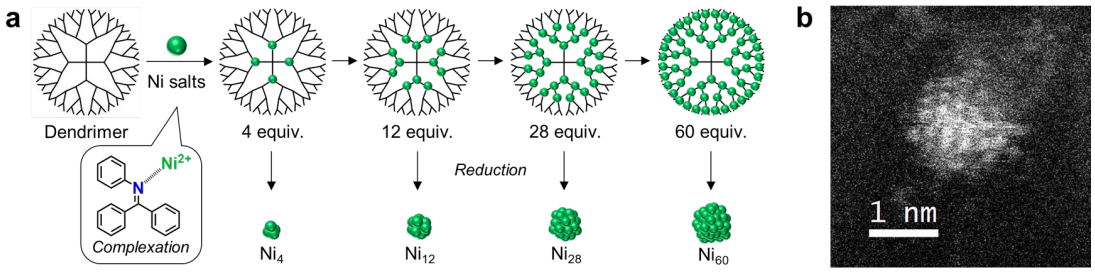

Fig. 1. **a** The scheme for stepwise complexation of Ni salts into a dendrimer and synthesis of 4 types of Ni SNPs. **b** A STEM image of Ni_{28} on graphene at atomic resolution.

1) T. Tsukamoto *et al*. *Nat. Commun.* **2018**, *9*, 3873. 2) T. Moriai *et al.*, *Angew. Chem. Int. Ed.* **2020**, *59*, 23051-23055.