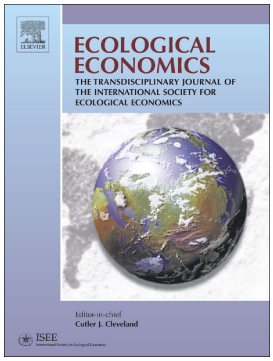

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Unexpected catalytic behavior of core-satellite gold nanostructures towards electroreduction of oxygen

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Abstract
Core-satellite gold nanoassemblies (CSNs) supported on carbon were fabricated by self-assembly using simple molecular cross-linkers and applied to the advanced oxygen reduction reaction (ORR). The as-prepared CSNs exhibited unexpectedly high catalytic activity and a dominant four-electron pathway toward the ORR.

Keywords
Core-satellite nanoassemblies; Molecular cross-linkers; Oxygen reduction reaction; Electrocatalysis; Fuel cell.

1. Introduction

Nanomaterials are used widely in a range of electrochemical applications, such as electrocatalysis, electrochemical analysis, and electrochemical synthesis, owing to their physical and chemical properties [1-3]. Versatile synthetic routes to nanomaterials have been developed to allow control of the morphology, structure and surface chemistry [4-8], which are crucial parameters in both fundamental studies and applications. Au together with other metals [9-11] and Au–metal alloy [12-14] nanostructures have attracted increasing attention for electrocatalytic applications, particularly in the field of fuel cells, because of their high chemical stability, catalytic properties, and easy surface modification [15-17]. In addition, the electrocatalytic activity of Au can be altered dramatically by surface activation with electrochemical [18-20], thermal [21-23], or chemical pretreatments [24,25]. On the other hand, gold itself is considered a poor catalyst for the oxygen reduction reaction (ORR) compared to other precious metals, such as Pd and Pt, due to the weak chemisorption properties caused by the filled d-band [26-29].

In the present study, a series of well-defined core-satellite Au nanoassemblies (CSNs) with different porosities, i.e. with the core-to-satellite gap distance controlled on a molecular scale, were synthesized. The effects of the interparticle distances on the ORR efficiency of the catalysts derived from the CSNs were investigated systematically by varying the length of the alkanedithiol molecular linkers ($HS(CH_2)_nSH$; n = 4, 8, 10). The series of as-prepared CSNs catalysts on carbon supports are denoted as CSNs(n)/C based on the length of the chain between the core and the satellites. This simple method is significant in preparing high-performance metal nanocatalysts with a well-controlled morphology, i.e. emphasizing the importance of tailored porosity in heterogeneous catalysis. Indeed, the electrocatalytic activity depends not only on the active site, but also on the surface morphology, particle sizes and surface area to volume ratio [30]. The catalytic ORR properties of single Au nanoparticles (GNs) of different sizes – 50 nm (core), 13 nm (satellite), and a 1:1 (v/v) mixture of 13 and 50 nm – are also given for comparison, and the corresponding products are called GNs(c), GNs(s), and GNs(m), respectively. Although